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ANALYTICAL STUDIES OF SPATIAL AND TEMPORAL CONFINEMENT IN STIMULATED RAMAN SCATTERING

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ABSTRACT

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The work presented in this thesis is a description of theoretical techniques for spatial and temporal confinement in the small signal regime of Stimulated Raman Scattering with a pump laser beam. The aim of this work is to provide where possible a mathematical model for the effects of confinement on both the pump, and the Raman generated Stokes fields, whilst at the same time to give some idea of the tools available to the theoretician persuing this end. Particular attention has been paid to the (existing) domains over which relatively simple mathematical models are applicable, and also to provide bounds on the applicability of both original and existing results.

Both the Maxwell and Lagrange formulation of the (electromagnetic) propagation problem are developed in this work. The paraxial ray equation which arises from the former is investigated in some detail; results are presented which give the full set of refractive index variations for which this equation is separable (and therefore potentially soluble) under an arbitrary transformation. The Lagrange formulation is employed to solve the spatial confinement problem which may arise from the use of a waveguide or a focussed pump beam. The traditional Maxwell formulation is used to provide the solutions to the temporal confinement problem. Where possible, results are presented which combine the solutions from both domains to obtain a model for simultaneous spatial and temporal confinement.

CONTENTS

Chapter 3

Raman Scattering in a Waveguide

Chapter 4

Raman Scattering with a Time Varying Pump Beam

Appendix 1

Separable Solutions To The Diffusion Equation

Appendix 2

Paper: Analysis Of Raman Gain With A Focussed Pump Beam

INTRODUCTION

Stimulated Raman Scattering may be regarded as a technique for conversion of energy from a pump to a Stokes field at a lower frequency. The conversion process is a result of an interaction of both fields with a Raman active medium through a third order non-linear susceptability. If the pump field is a monochromatic plane wave, then under the appropriate conditions, there will be an exponential growth - at a rate proportional to the intensity of the pump field - of a co-propagating, co-linearly polarised field at the Stokes frequency.

Often, it is required to maximise the efficiency of the conversion process perhaps to produce a (new) high intensity coherent source of radiation at a longer wavelength with minimum 'cost' in terms of the source radiation. Thus, for a continuous plane-wave pump field, there is an increase in the conversion efficiency with pump intensity, so that given a coherent source at some fixed *power*, one may be tempted to increase the efficiency by confining this power to a smaller cross sectional area - i.e. by focussing the pump beam. However, one can no longer expect the simple dependence of exponentional growth on pump intensity to hold; spatial confinement will cause both pump and generated Stokes beams to diffract thereby reducing the volume over which the maximal growth rate can take place.

Similarly, given a pulsed source at some fixed *energy*, one may attempt to increase the conversion efficiency by confining the energy to a smaller pulse width and thereby increasing the effective intensity at the pump frequency. However, once again we must consider aspects of the conversion process which destroy the simple relationship between exponential growth rate and pump intensity. Once the pump pulse becomes sufficiently short, it may no longer be regarded as a *continuous* plane wave; the finite response time of the Raman medium will impede the conversion process so that a steady-state is never attained.

For both of the suggestions above there is clearly *some* advantage in attempting to increase the conversion efficiency through spatial and temporal confinement, although the degree to which this is true is not apparent from the outline above. The goal of this thesis is to quantify the effects of confinement so that the efficacy of the various techniques can be evaluated, and the experimentally adjustable parameters thereby chosen to give a Stokes field with the desired characteristics.

Chapter 1 may be regarded as an introduction to the more detailed theoretical deliberations of chapters 2 to 4. The material of the constituent sections has been arranged to provide a linear progression from the general Lagrangian formulation for the Electromagnetic fields, to the theory of small-signal steady-state plane-wave Stimulated Raman Scattering. The intervening material contains explanations of the various approximations - which are often implicit in the literature - associated with the behaviour of electromagnetic fields in inhomogeneous, anisotropic media.

In Chapter 2, there is a theoretical analysis of the effects of pump focussing on the conversion process. The results are derived from a Lagrangian formulation of the small-signal steady-state problem of Raman Scattering with a Gaussian pump beam. Using this approach it has been possible to obtain an analytic description of the Stokes field which is shown to be in very close agreement with an exact numerical treatment, and which also conforms to the established behaviour in the limiting cases of high and low pump power. The results of this chapter are also presented in the form of a paper in Appendix 2.

Chapter 3 contains a theoretical analysis of the effects of a capillary waveguide on the Raman Scattering process. This time the spatial confinement is achieved by virtue of the reflections of the pump and Stokes beams at the waveguide walls. Just as in free-space Raman Scattering, there are aspects of the conversion process that destroy the simple dependency of Stokes growth rate on pump intensity. In this case, one must consider in particular the effects of losses on reflection at the waveguide walls which become more severe as the bore of the guide is decreased. Particular attention in this chapter is given to the correct choice of design parameters which maximise the efficiency of the conversion.

Chapter 4 contains a review of the theory of Stimulated Raman Scattering using a plane-wave, time-varying pump field. This theory is then applied to the analysis of Raman Scattering with a dual mode pump laser. In this chapter it is also shown how the plane wave results can, in some circumstances, be generalised to include the effects of both temporal confinement, and the spatial confinement techniques considered in chapters 2 and 3.

Appendix 1 is an exposition of the theory of separable solutions to the diffusion equation. This work evolved from earlier attempts to find exact solutions to the gain-focussing problem discussed in chapter 2, and was subsequently developed to cover all possible refractive index variations. It is believed that the results may find a general utility in optics and more generally to physical systems which can be described by a diffusion equation (e.g. quantum mechanics and thermodynamics).

CHAPTER 1

ELECTROMAGNETIC FIELDS IN A POLARISABLE MEDIUM

1. Introduction

In this chapter we discuss the electromagnetic field equations applicable to the propagation of a field in an electrically polarisable medium. The aim is to derive a set of equations which are easily adapted to the context of each of the later chapters in this thesis.

Of particular interest to us in this chapter are the formulations of both the classical Lagrangian and the Maxwell equations. For each of these formulations we discuss the zero divergence approximation, the slowly varying envelope approximation, and the paraxial-ray approximation, each of which is employed at some point in the work which follows. These approximations are brought together in the last section to give the standard result for small-signal, steady-state, plane-wave Raman Scattering, against which the results for the various techniques for spatial and temporal confinement can be compared.

2. Classical formulation

The starting point for this chapter is the electromagnetic Lagrangian density (Goldstein, 1950):

$$\mathcal{L} = \tfrac{1}{2}\,[\underline{D}.\underline{E} - \underline{B}.\underline{H}] \qquad [1]$$

where each of the symbols have their usual meaning. It is to be understood that the correct description of the system is found by choosing fields that minimise the Lagrangian:

$$L = \iiiint \mathcal{L} \; d\Omega.dt \qquad [2]$$

where the volume and time integrations are over the region (Ω,t) for which the fields are to be determined. Throughout this thesis, the reaction of the medium $\underline{D}$, $\underline{B}$ to the electromagnetic fields $\underline{E}$, $\underline{H}$ can be described by the relations:

$$\underline{D} = \epsilon_0\underline{E} + \underline{P} \qquad [3]$$

and

$$\underline{B} = \mu_0\underline{H} \qquad [4]$$

where $\underline{\nabla} \times \underline{E} = -\delta\underline{B}/\delta t$ is understood. [5]

i.e. we consider media that are electrically polarisable only. Also, we will deal with polarisations such that it will always be possible to write:

$$\underline{\nabla}.\underline{D} = 0 \qquad [6]$$

$$\underline{\nabla}.\underline{B} = 0 \qquad [7]$$

which can be interpreted as the absence of free charge (rather than radiating dipoles).

The polarisation $\underline{P}$ will, in general, depend on the spatially varying characteristics of the medium. In the case of a linear optical system employing lenses and mirrors, the character of $\underline{P}$ is determined by the refractive index of the medium, i.e.:

$$\underline{P} = \epsilon_o \chi(\underline{r})\underline{E} \qquad [8]$$

$$X(\underline{r}) = n^2(\underline{r}) - 1 \qquad [9]$$

Where $\chi(\underline{r})$ is the linear susceptibility, and $n(\underline{r})$ the refractive index; $\chi(\underline{r})$ is of course a scalar if the medium is isotropic. More generally however, the polarisation of the medium depends in a non-linear fashion, on the propagating fields, and through this the susceptibility may become inhomogeneous, anisotropic and time dependent (see section 3).

The relations [3]-[7] can be employed to cast [1] solely in terms of the electric field and the polarisation. It is convenient however, to follow traditional methods and define a related quantity $\underline{A}$ where:

$$-\delta\underline{A}/\delta t = \underline{E} . \qquad [10]$$

Choosing the Coulomb gauge, it follows from [5] and [7] that:

$$\underline{B} = \underline{\nabla} X \underline{A} . \qquad [11]$$

With these substitutions, [1] becomes:

$$\mathcal{L} = -1/(2\mu_o)[(\underline{\nabla} X \underline{A})^2 - (1/c)^2.(\delta\underline{A}/\delta t)^2 + (\delta\underline{A}/\delta t)\underline{P}/(c^2\epsilon_o)] \qquad [12]$$

The first two terms alone describe the space and time behaviour of the field in a vacuum. The last term in [12] describes the interaction of the field $\underline{A}$ with the polarisation of the medium $\underline{P}$. Note that an alternative, form for [12] can be obtained by using the identity:

$$(\underline{\nabla}\times\underline{A})^2 = \left(\frac{\partial\underline{A}}{\partial x}\right)^2 + \left(\frac{\partial\underline{A}}{\partial y}\right)^2 + \left(\frac{\partial\underline{A}}{\partial z}\right)^2 - \left(\underline{\nabla}\,\underline{A}\right)^2 + 2\left[\frac{\partial A_x}{\partial x}\frac{\partial A_z}{\partial z} - \frac{\partial A_x}{\partial z}\frac{\partial A_z}{\partial x}\right]$$
$$+ 2\left[\frac{\partial A_x}{\partial x}\frac{\partial A_y}{\partial y} - \frac{\partial A_x}{\partial y}\frac{\partial A_y}{\partial x}\right] + 2\left[\frac{\partial A_y}{\partial y}\frac{\partial A_z}{\partial z} - \frac{\partial A_y}{\partial z}\frac{\partial A_z}{\partial y}\right] \qquad [13]$$

2.1 Boundary conditions

Often we find that the electric field is given on the boundary $z = 0$, whilst it is kown that the fields vanish for all z as $x,y \to \infty$. There are of course, problems in being able to specify exactly such boundary conditions, but at this point it is convenient to assume that they are not insurmountable and therefore that

$$\underline{A}(x,y,z,t) \text{ at } z = 0 \text{ is given;} \qquad [14]$$

and $$\mathrm{Lt}_{x,y\to\infty}\ \underline{A}(x,y,z,t) = 0. \qquad [15]$$

In addition, it can usually be assumed that the plane $z = 0$ is perpendicular to the 'direction of propagation' of the electric field. Strictly, this concept is antithetical to that of a boundary condition in that it arises only in the description of a field over a finite extent outside the active medium. However, it can be interpreted as requiring that the main spatial variation of the field determines the boundary condition:

$$\delta/\delta z\ \underline{A}(x,y,z,t) \text{ at } z = 0 \text{ is given.} \qquad [16]$$

Under the Coulomb gauge, this means the longitudinal component of the electric field is small compared to the transverse components, and that:

$$A_z(x,y,z,t) \simeq 0 \text{ at } z = 0. \qquad [17]$$

With the qualifications of sections 4 and 6, we can conclude that the longitudinal component of the electric field is everywhere negligibly small. This conclusion, along with [15], requires the last three terms in [13] vanish on integration over the volume in [2]. Therefore, they may be ignored and the effective Lagrangian density written:

$$\mathcal{L} = -\frac{1}{2\mu_0}\left[\left(\frac{\partial \underline{A}}{\partial x}\right)^2 + \left(\frac{\partial \underline{A}}{\partial y}\right)^2 + \left(\frac{\partial \underline{A}}{\partial z}\right)^2 - \frac{1}{c^2}\left(\frac{\partial \underline{A}}{\partial t}\right)^2 - \left(\underline{\nabla}\,\underline{A}\right)^2 + \frac{\underline{P}}{\varepsilon_0 c^2}\frac{\partial \underline{A}}{\partial t}\right] \qquad [18]$$

which is the required result for the electro-kinetic potential in an electrically polarisable medium.

2.2 The Euler-Lagrange equations

The equivalent Maxwell formulation is obtained by finding the field $\underline{A}(x,y,z,t)$ that satisfies the Euler-Lagrange equations for the problem:

$$\delta\left\{\iiiint \mathcal{L}\left(\underline{r}, t, \frac{\partial \underline{A}}{\partial x}, \ldots\right) dx\,dy\,dz\,dt\right\} = 0 \qquad [19]$$

Hence, we must have for each $\alpha \in \{x,y,z,t\}$

$$\left\{\frac{\partial}{\partial A_\alpha} - \frac{d}{dx}\frac{\partial}{\partial\, \partial A_\alpha/\partial x} - \frac{d}{dy}\frac{\partial}{\partial\, \partial A_\alpha/\partial y} - \frac{d}{dz}\frac{\partial}{\partial\, \partial A_\alpha/\partial z} - \frac{d}{dt}\frac{\partial}{\partial\, \partial A_\alpha/\partial t}\right\}\mathcal{L} = 0 \qquad [20]$$

which from [18] gives:

$$\square A_\alpha + \frac{1}{2\varepsilon_0 c^2}\sum_\beta \frac{d}{d\beta}\left[\frac{\partial \underline{P}}{\partial\, \partial A_\alpha/\partial\beta}\cdot\frac{\partial \underline{A}}{\partial t}\right] - \frac{d}{d\alpha}\left(\underline{\nabla}\,\underline{A}\right)$$

$$+ \frac{1}{2\varepsilon_0 c^2}\left[\frac{\partial P_\alpha}{\partial t} - \frac{\partial \underline{P}}{\partial A_\alpha}\cdot\frac{\partial \underline{A}}{\partial t}\right] = 0 \qquad [21]$$

where $\beta \in \{x,y,z,t\}$ and $\square$ is the D'Alembertian operator. For the terms above involving the polarisation, we have adopted the convention that the total derivatives act on both the field variables and the co-ordinates, whilst the partial derivatives act only on one or the other as indicated.

3. Linear polarisation

Of particular interest in this thesis is a polarisation that is linear in the electric field, but not necessarily homogeneous or isotropic. A generalisation of [8] would then be:

$$\underline{P} = \epsilon_o \underline{\underline{\chi}}(\underline{r})\underline{E} \qquad [22]$$

where $\underline{\underline{\chi}}(\underline{r})$ is a 3x3 matrix. This form for the polarisation can arise for instance in small signal Stimulated Raman Scattering when the pump field is approximately unperturbed by the growth of the Stokes field and can therefore be regarded as given. In this case the polarisation at the Stokes frequency can be written:

$$\underline{P}_s = \epsilon_o(n_s^2-1)\underline{E}_s + (3/2)\epsilon_o\chi(-\omega_s;\omega_p,-\omega_p,\omega_s)\underline{E}_p\underline{E}_p{}^*\underline{E}_s \qquad [23]$$

whilst the polarisation at the pump frequency is just:

$$\underline{P}_p = \epsilon_o(n_p^2-1)\underline{E}_p \qquad [24]$$

where $\underline{E}_p$ and $\underline{E}_s$ denote the complex amplitudes of the Fourier exponentials for the pump and Stokes fields respectively, i.e.

$$\underline{E} = \mathrm{Re}\ \{\underline{E}_s\exp(i\omega_s t) + \underline{E}_p\exp(i\omega_p t)\}$$

and $\chi(-\omega_s;\omega_p,-\omega_p,\omega_s)$ is the (2nd rank) tensor for Stimulated Raman Scattering, (the coefficient 3/2 is chosen to be consistent with Hanna et al, 1979). In order to recover a form similar to that of [22], we must now borrow from the work of Yuratich and Hanna (1977) which deals with the susceptibility tensor for coherent anti-Stokes Raman Scattering. In that work we must interpret the angular factor (see equation 15b), which relates the polarisations of the Stokes, pump and anti-Stokes fields, as applying instead to *Stimulated* Raman Scattering where now the anti-Stokes field is replaced by a 'Scattered' Stokes field:

$$\underline{P}_s = \epsilon_o(n_s^2-1)\underline{E}_s + (3/2)\epsilon_o|E_p|^2\underline{\underline{\chi}}^{(R)}\underline{E}_s \qquad [25]$$

where $\underline{\underline{\chi}}^{(R)}$ is the orientation averaged Raman Scattering matrix and is in general complex. Thus we recover the form [22] in which the effective linear susceptibility is:

$$\underline{\underline{\chi}}(\mathbf{r}) \quad = \quad n_s^2-1 \;+\; (3/2)|E_p|^2\underline{\underline{\chi}}^{(R)} \qquad [26]$$

The scattering matrix $\underline{\underline{\chi}}^{(R)}$ can be calculated from the direction of the pump polarisation, and the isotropic, and the anisotropic (symmetric and antisymmetric) parts of the Raman polarisability tensor, denoted here by α^2, β^2 and γ^2 respectively. Thus if the pump field is linearly polarised in the x direction we find:

$$\underline{\underline{\chi}}^{(R)} \quad \alpha \quad \begin{bmatrix} \alpha^2+4\beta^2/45, & 0 & , & 0 \\ 0 & ,\beta^2/15+\gamma^2/9, & & 0 \\ 0 & , & 0 & ,\beta^2/15+\gamma^2/9 \end{bmatrix} \qquad [27]$$

It follows from the above that the susceptibility $\underline{\underline{\chi}}^{(R)}$ can be written in terms of the 'depolarisation ratio' ρ which is the ratio of intensities scattered at the Stokes frequency (per unit solid angle) in the directions colinear, and perpendicular to the incident (pump) radiation. This quantity is found to be (Yuratich and Hanna, 1977):

$$\rho \quad = \quad (3\beta^2+5\gamma^2)/(45\alpha^2+4\beta^2) \qquad [28]$$

Note that whilst the definition of ρ applies only to spontaneous Raman Scattering, the quantity is still useful in defining a general susceptibility for Stimulated Raman Scattering:

$$\chi^{(R)} \quad = \quad \chi^{(R)}_{xx} \begin{bmatrix} 1 \;, & 0 \;, & 0 \\ 0 \;, & \rho \;, & 0 \\ 0 \;, & 0 \;, & \rho \end{bmatrix} \qquad [29]$$

If instead the pump field is circularly polarised:

$$\mathbf{E}_p \quad = \quad E_p(\mathbf{e}_x \;+\; i\mathbf{e}_y)/\sqrt{2} \qquad [30]$$

then the susceptibility matrix is unchanged from that above provided one interprets the vector components denoting the Stokes polarisation in a spherical polar coordinate system. Alternatively, in a Cartesian coordinate system, the scattering matrix for circularly polarised pump radiation undergoes a similarity transform and thereby becomes:

$$\underline{\underline{\chi}}^{(R)} = \chi^{(R)}_{xx} \begin{bmatrix} (1+\rho)/2 \;, & (1-\rho)/2 \;, & 0 \\ (1-\rho)/2 \;, & (1+\rho)/2 \;, & 0 \\ 0 \;, & 0 \;, & \rho \end{bmatrix} \qquad [31]$$

The value of ρ depends on the symmetry of the scattering molecule and generally is much less than one. For the cases considered in this thesis, prompted by the experimental work on Hydrogen and Methane gases, we are justified in neglecting the component of the Stokes field polarised orthogonally to that of the pump. (Note that a small difference between the diagonal elements in [29] is exponentiated by the gain process in the classical plane wave - small signal, steady state - formulation of Raman Scattering.) In the appropriate coordinate system then, $\underline{\underline{\chi}}^{(R)}$ is a diagonal matrix with one element significantly larger than the other two. The implication for the field equations is that, provided the small divergence approximation holds (see section 4), then generally it is sufficient to concentrate on the behaviour of the component of the Stokes electric field vector work which is colinear with the pump polarisation. (This is the policy which has been adopted in the later chapters.)

For the given linear form for the polarisation, we can now determine the full dependence of the Lagrangian density on the various components of the vector field. Thus [18] is found to be:

$$\mathcal{L} = -\frac{1}{2\mu_0}\left[\sum_\alpha \left(\frac{\partial \underline{A}}{\partial \alpha}\right)^2 - \left(\underline{\nabla}\cdot\underline{A}\right)^2 - \frac{\partial \underline{A}^T}{\partial t}\, \frac{\underline{\underline{\chi}}(\underline{r})}{c^2}\, \frac{\partial \underline{A}}{\partial t} \right] \qquad [32]$$

and [19] becomes, for each $\alpha \in \{x,y,z\}$:

$$\Box E_\alpha - \frac{\hat{\underline{\alpha}}\; \underline{\underline{\chi}}(\underline{r})}{c^2}\frac{\partial^2 \underline{E}}{\partial t^2} - \frac{d}{d\alpha}\left(\underline{\nabla}\cdot\underline{E}\right) = 0 \qquad [33]$$

which can be written collectively:

$$\Box \underline{E} = \underline{\nabla}\left(\underline{\nabla}\,\underline{E}\right) + \frac{\underline{\underline{\chi}}(\underline{r})}{c^2}\frac{\partial^2 \underline{E}}{\partial t^2} \qquad [34]$$

which is the standard form for the Maxwell equations in a polarisable medium. Note that [34] no longer holds if the susceptibility is a function of time or if the linearity condition expressed by [22] is violated.

4. Small divergence approximation

Without the divergence term, [34] is a wave equation for the electric field. That the term vanishes when the medium is homogeneous (though not necessarily isotropic) can be seen from [6] and [22]. When this is not the case, it may be convenient to express the divergence using the relative permittivity tensor as follows:

$$\underline{\nabla}\,\underline{E} = \left[\frac{\partial}{\partial\alpha}\left(\epsilon^{-1}\right)_{\alpha,\beta}\right]\epsilon_{\beta,\gamma}\,E_{\gamma} \qquad [35]$$

where $\epsilon_{\alpha,\beta} = \delta_{\alpha,\beta} + \left[\chi(\underline{r})\right]_{\alpha,\beta}$

and $\alpha, \beta, \gamma \in \{x, y, z\}$

and summation over repeated indices is implied. The functional behaviour of the susceptibility determines the conditions under which [35] can be neglected.

A strong motivation for ignoring the divergence term is that the resulting equations for the electric field vector are greatly simplified. In particular, if the susceptibility is diagonal, then [34] decouples into three independent wave equations. Generally a sufficient condition for neglecting the field divergence is that the fractional change of the total refractive index tensor be small within a 'characteristic distance' of the electric field. In most cases (see section 6) this characteristic distance is simply the wavelength of the field along the axis of propagation (see for example Yariv, 1975). If a method of validating this approximation is required, it is suggested that a basic test for consistency would be to compare the resulting magnitude of the divergence term with the other terms in [34].

4.1 Waveguides

If the field vector is confined by a waveguide it may no longer be possible to assume that the divergence terms can be neglected. In particular, the approximation discussed above clearly breaks down if there is a step discontinuity in the refractive index at the waveguide wall. In these cases, it is usual to assume zero divergence of the field either side of, but not over, the wall. Continuity of the fields over the wall will then determine the relation between the core and cladding fields. (This is the approach adopted in chapter 3).

5. The slowly varying envelope approximation

We now consider the consequences of assuming that the temporal behaviour of the electric field is primarily that of a single Fourier component, i.e. that:

$$\underline{A}(\underline{r},t) = \mathrm{Re}\left\{ \underline{a}(\underline{r},t)\, e^{i\omega t} \right\} \qquad [36]$$

where $\underline{a}(\underline{r},t)$ is a 'slowly varying' field. For this decomposition to have any meaning, it is clear that the magnitude of the Fourier transform of the field $\underline{a}(\underline{r},t)$ must be confined to a band that lies well within the interval $(0,\omega)$. We can then capitalise on this situation by approximating the time derivative in [18]. The argument runs as follows:
Since

$$\left(\frac{\partial \underline{A}}{\partial t}\right)^2 = \frac{1}{4} e^{2i\omega t}\left[i\omega \underline{a} + \frac{\partial \underline{a}}{\partial t}\right]^2 + \frac{1}{4} e^{-2i\omega t}\left[-i\omega \underline{a}^* + \frac{\partial \underline{a}^*}{\partial t}\right] + \frac{1}{2}\left| i\omega \underline{a} + \frac{\partial \underline{a}}{\partial t}\right|^2 \qquad [37]$$

then provided

$$\left|\frac{\partial \underline{a}}{\partial t}\right|^2 << \omega^2 |\underline{a}|^2 \qquad [38]$$

the second order terms in the time derivative of $\underline{a}(\underline{r},t)$ can be neglected. Moreover, the first and second terms in [38] will contribute a negligible amount to the Lagrangian [2] by virtue of the phase cancallation of the sinusoid components (see for example Loudon, 1968). That this is true follows from consideration of the integral:

$$\int_{-\infty}^{\infty} \exp\left[i\omega t - t^2/\tau^2\right] dt = \sqrt{\pi}\exp\left[-\omega^2\tau^2/4\right] \qquad [39]$$

And this result must be compared with that for $\omega = 0$:

$$\left| \frac{\int_{-\infty}^{\infty} \exp\left[i\omega t - t^2/\tau^2\right] dt}{\int_{-\infty}^{\infty} \exp\left[-t^2/\tau^2\right] dt} \right|^2 = \exp\left[-\frac{\omega^2\tau^2}{2}\right] \qquad [40]$$

Therefore if the field envelope is taken to be a Gaussian of (temporal) width τ,

$$\underline{a} \sim \exp\left[-t^2/\tau^2\right]$$

then the slowly varying envelope approximation [38] requires that:

$$\omega^2\tau^2 \gg 4\, t^2/\tau^2 \qquad [41]$$

The worst case is towards the end of the pulse when $t/\tau \sim 1$ and so by virtue of [40], the rapidly varying terms in [37] are seen to be averaged out of the Lagrangian. It follows from this argument that [37] can be simplified to:

$$\left(\frac{\partial \underline{A}}{\partial t}\right)^2 \rightarrow \frac{-\omega^2|\underline{a}|^2}{4} + \frac{i\omega}{2}\left[\underline{a}\frac{\partial \underline{a}^*}{\partial t} - \underline{a}^*\frac{\partial \underline{a}}{\partial t}\right] \qquad [42]$$

and further, that only the time averaged components of the remaining terms in [18] need be retained (Loudon, 1968), i.e.

$$\left(\frac{\partial \underline{A}}{\partial \alpha}\right)^2 \rightarrow \frac{1}{2}\left|\frac{\partial \underline{a}}{\partial \alpha}\right|^2 \qquad [43]$$

Under the same approximation, the Maxwell equations [21] are modified as follows:

$$\begin{aligned}
&\nabla^2 a_\alpha + \frac{\omega^2}{c^2}a_\alpha - 2i\omega\frac{\partial a_\alpha}{\partial t} - \frac{\partial}{\partial \alpha}\left(\underline{\nabla}.\underline{a}\right) \\
&+ \frac{1}{2\varepsilon_0 c^2}\sum_\beta \frac{d}{d\beta}\left[\frac{\partial P}{\partial\, \partial a_\alpha/\partial\beta}\left(\frac{\partial \underline{a}}{\partial t} + i\omega\underline{a}\right)\right] \\
&+ \frac{1}{2\varepsilon_0 c^2}\left[e^{-i\omega t}\frac{\partial P_\alpha}{\partial t} - \frac{\partial P}{\partial a_\alpha}\left(\frac{\partial \underline{a}}{\partial t} + i\omega\underline{a}\right)\right] \\
&+ \left[\frac{d}{dt}\frac{\partial P}{\partial\, \partial A_\alpha/\partial t}\right].\left[\frac{\partial \underline{a}}{\partial t} + i\omega\underline{a}\right] + \frac{\partial P}{\partial\, \partial A_\alpha/\partial t}\left[-\omega^2\underline{a} + 2i\omega\frac{\partial \underline{a}}{\partial t}\right] = 0
\end{aligned} \qquad [44]$$

where now $\beta \in \{x,y,z\}$ only, and it is to be understood that $a_\alpha(\underline{r},t)$ is evaluated as [36].

The condition in the Maxwell formulation that is equivalent to [38] is thus seen to be:

$$\left| \frac{\partial^2 \underline{a}}{\partial t} \right| << 2\omega \left| \frac{\partial \underline{a}}{\partial t} \right| \qquad [45]$$

Equation [44] can simplify considerably if assumptions are made about the relation between the polarisation $\underline{P}$ and the field $\underline{A}$. Without further information it is generally more convenient to work with the more compact Lagrangian formulation.

5.1 Justification

So far we have discussed only the requirements for validity of the slowly varying envelope approximation within the region of integration of [2]. In terms of the electric field propagating into this region from the boundary, a sufficient condition is that the boundary field itself satisfies [38], whilst the time dependent characteristics of the medium, through the susceptibility, be negligible over the period $2\pi/\omega$. Support for the first of these conditions comes from consideration of the frequencies and pulse widths encountered in non-linear optics. For instance, a He-Ne Laser (λ = 633nm) in a cavity of length l = 30cm will generate pulses with:

$$\omega^2\tau^2/4 \simeq (\pi l/\lambda)^2 \simeq 10^{12} \qquad [46]$$

A similar calculation shows that the second condition is also easily satisfied. For instance, Raman Scattering of a laser (λ = 532nm) in H_2 gas has a bandwidth of $\Gamma \simeq$ 720MHz at 20 atm. (Pratt, 1985). This determines a 'lifetime' Γ^{-1} for the process which can be taken as the period τ in [41], whence:

$$\omega^2\tau^2/4 \simeq 5.10^{12} \qquad [47]$$

(Note that for the generated Stokes component to be relatively speaking 'steady state' the requirement is more stringent - see chapter 4). In practice then, we are justified in applying the slowly varying envelope approximation and [38] can be used to monitor the consistency in terms of the field envelopes. We note finally that in the absence of a dynamical variation in the susceptibility, the single Fourier component is an eigenfunction of the separated wave equation [21]. Therefore there is no approximation in using the results of this section with $\delta/\delta t\ \mathbf{a}(\mathbf{r},t) = 0$.

6. The paraxial approximation

This approximation is analagous to that of the previous section with spatial replacing temporal considerations. The difference here is that the paraxial approximation is usually taken to be a 'package' wherein the field varies predominantly in the lonitudinal direction, but also in which the transverse spatial derivatives cannot be ignored (i.e. the fields are not plane-wave). As before, the field is expanded in terms of slowly varying envelopes and single Fourier components:

$$\underline{A}(\underline{r},t) = \mathrm{Re}\left\{ \underline{a}(\underline{r},t)\, e^{-ikz} \right\} \qquad [48]$$

where the $\underline{a}(\underline{r},t)$ is slowly varying in the z direction, the spatial Fourier transform of which is well confined in the interval $(0,\underline{k})$. For definiteness, it is usually assumed that the sinusoid behaviour of the field occurs in the z (or longitudinal) direction (see section 2.1); thus in combination with the expansion [36] for the time varying component, [48] describes a propagating wave with speed ω/k in the z direction.

Following the analysis of section 5 we have :

$$\left| \frac{\partial \underline{a}}{\partial z} \right|^2 << k^2 |\underline{a}|^2 \qquad [49]$$

$$\left(\frac{\partial \underline{A}}{\partial z} \right)^2 \longrightarrow -\frac{k^2 |\underline{a}|^2}{4} - \frac{ik}{2} \left[\underline{a} \frac{\partial \underline{a}^*}{\partial z} - \underline{a}^* \frac{\partial \underline{a}}{\partial z} \right] \qquad [50]$$

whilst remaining terms for $\alpha \in \{x,y,t\}$ become:

$$\left(\frac{\partial \underline{A}}{\partial \alpha} \right)^2 \longrightarrow \frac{1}{2} \left| \frac{\partial \underline{a}}{\partial \alpha} \right|^2 \qquad [51]$$

A result analagous to [43] follows for the Maxwell equations for which we obtain the constraint equivalent to [49] (see for example, Yariv 1975):

$$\left| \frac{\partial^2 \underline{a}}{\partial z^2} \right| << 2k \left| \frac{\partial \underline{a}}{\partial z} \right| \qquad [52]$$

6.1 Justification

The justification for the paraxial approximation depends on both the boundary conditions on the electric field and the variation of the susceptibility in the z direction. If the induced longitudinal behaviour of the electric field inside the region Ω is seen to derive primarily from that of the 'external field' impinging on the boundary $z = 0$, then the justification for the validity of the first condition remains unchanged from that of the previous section.

The second condition can be supported once again with an example from Raman Scattering of a laser ($\lambda_p = 532\text{nm}$) in H_2 gas. Under appropriate conditions for the pump field (see section 7), the Stimulated Raman Scattering plane-wave gain coefficient may have a value $G_R \simeq 30$ whereby the intensity at the Stokes wavelength ($\lambda_s = 681\text{nm}$) grows as $\exp(G_R z)$. Thus by analogy with [41] we have:

$$k_s^2/G_R^2 \simeq 9.10^{10} \qquad [53]$$

which in this case justifies the use of the paraxial approximation.

6.2 Waveguides

The parallel status of this approximation with the slowly varying envelope is destroyed if there are step discontinuities in the refractive index which occur for instance within a waveguide. Waveguides are employed to confine the electromagnetic field and are thus an important tool in the field of non-linear optics. A fuller discussion of their use is given in chapter 3. It is remarked at this point however, that although [46] may be satisfied, the paraxial approximation will generally be inappropriate for a waveguide field. This is because the mode structure of a waveguide is determined by the reflection/interference effects of the fields at the waveguide walls and therefore it is important to choose a coordinate system which simplifies the boundary conditions. Put another way, the characteristics of the Helmholtz equation can be made to coincide with the walls of a cylindrical waveguide, whereas the same is not generally true for the characteristics of the paraxial ray equation. There are cases however, where the characteristics of the paraxial ray equation can be made to lie along the waveguide walls (see appendix 1), as for instance is possible for a linearly tapered guide (truncated cone).

7. Plane Wave Raman Scattering

In this section we bring together some of the approximations described in this chapter which are pertinent to small-signal, steady-state, Raman Scattering of a plane-wave pump field. Our aim is to derive an expression for the evolution of a Stokes field under these conditions. The result acts as a reference against which the techniques dicussed in the later chapters of this thesis can be compared.

The polarisation induced by a pump field at the Stokes frequency in the small-signal, steady-state regime has been given in section 3:

$$\underline{P}_s = \varepsilon_0 (n_s^2 - 1)\underline{E}_s + (3/2)\,\varepsilon_0 \underline{\underline{\chi}}^{(R)} \underline{E}_s |E_p|^2 \qquad [25]$$

In we interpret this as a linear polarisation at the Stokes frequency, then we can employ the Maxwell equations of that section (equation [34]) with the small divergence approximation of section 4 to give:

$$\left[\nabla^2 - \frac{1}{c^2}\left[n_s^2 + 3/2\,|E_p|^2 \underline{\underline{\chi}}^{(R)} \right] \frac{\partial^2}{\partial t^2} \right] \underline{E} = 0 \qquad [54]$$

corresponding to a Lagrangian density (from [32]):

$$\mathcal{L} = \frac{-1}{2\mu_0}\left[\sum_\alpha \left(\frac{\partial \underline{A}}{\partial \alpha} \right)^2 - \frac{\partial \underline{A}^T}{\partial t} \frac{\underline{\underline{\chi}}^{(R)}}{c^2} \frac{\partial \underline{A}}{\partial t} \right] \qquad [55]$$

Also in section 3, we have argued that we need generally only consider the component of the Stokes field which has a polarisation which is colinear with that of the pump. Then the above becomes:

$$\left[\nabla^2 - \frac{1}{c^2}\left[n_s^2 + 3/2\,|E_p|^2 \chi^{(R)}_{xx} \right] \frac{\partial^2}{\partial t^2} \right] E_x = 0 \qquad [56]$$

with Lagrangian density:

$$\mathcal{L} = \frac{-1}{2\mu_0}\left[(\underline{\nabla} A_x)^2 - \left(\frac{\partial A_x}{\partial t} \right)^2 \frac{1}{c^2}\left[n_s^2 + 3/2\,|E_p|^2 \chi^{(R)}_{xx} \right] \right] \qquad [57]$$

where we have assumed a pump polarised in the x direction for definiteness. If the pump intensity is constant in time, then the above admits of solutions:

$$\underline{E} = \mathrm{Re}\left\{ \underline{E}_s \exp\left[i\omega_s t \right] \right\} \qquad [58]$$

whereupon [56] becomes $\left(\kappa_s^2 = n_s^2 \omega_s^2 / c^2 \right)$:

$$\left[\nabla^2 + \kappa_s^2 + 3/2\, \kappa_s^2 / n_s^2 \left| E_p \right|^2 \chi_{xx}^{(R)} \right] E_{sx} = 0 \qquad [59]$$

Generally the magnitude of the 3rd term in braces is much less than the second, i.e. the magnitude of the pump induced polarisation is small compared to that of the linear polarisation of the medium. Therefore, whatever the spatial variation of the pump intensity, we can be sure of the justification for making the paraxial approximation (section 6). Thus assuming a propagation principally in the z direction, and a Stokes field polarised in the x direction:

$$E_{sx} = \epsilon_s(\underline{r}) \exp\left[-i\kappa_s z \right] \qquad [60]$$

equation [59] becomes:

$$\left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2i\kappa_s \frac{\partial}{\partial z} + 3/2\, \kappa_s^2 / n_s^2 \left| E_p \right|^2 \chi_{xx}^{(R)} \right] \epsilon_s = 0 \qquad [61]$$

and recalling the relation [10] between $\underline{A}$ and $\underline{E}$, the Lagrangian density is:

$$\mathcal{L} = \frac{-1}{4\mu_0\omega^2} \left[\left| \frac{\partial \epsilon_s}{\partial x} \right|^2 + \left| \frac{\partial \epsilon_s}{\partial y} \right|^2 - 2\kappa_s \mathrm{Im}\left\{ \epsilon_s^* \frac{\partial \epsilon_s}{\partial z} \right\} - \frac{3}{2} \frac{\kappa_s^2}{n_s^2} \left| E_p \right|^2 \chi_{xx}^{(R)} \left| \epsilon_s \right|^2 \right] \qquad [62]$$

The purpose of this section is to determine the evolution of a Stokes field when the pump is a plane wave. With this in mind, we are free to consider the evolution of a single Fourier component of the Stokes field whereupon the transverse derivatives above can be ignored:

$$\frac{\partial \epsilon_s}{\partial z} = -\frac{3i}{4} \frac{\kappa_s}{n_s^2} \chi_{xx}^{(R)} \left| E_p \right|^2 \epsilon_s \qquad [63]$$

The Stokes intensity therefore grows exponentially:

$$I_s(z) = I_s(0) \exp\left[G_R z \right] \qquad [64]$$

where $$G_R = \tfrac{3}{2}\, \mathrm{Im}\left\{\chi_{xx}^{(R)}\right\} k_s / n_s^2 \, |E_p|^2 \qquad [65]$$

is the small-signal, steady-state, plane-wave Raman gain coefficient. In addition it is often useful to refer to a Raman gain coefficient g_r where

$$g_r = 3\, \mathrm{Im}\left\{\chi_{xx}^{(R)}\right\} k_s \cdot n_p / n_s^2 \cdot 1/\varepsilon_0 c \qquad [66]$$

so that the Stokes intensity can be written:

$$I_s(z) = I_s(0) \exp\left[g_r I_p z \right] \qquad [67]$$

In this chapter then, we have confirmed the exponential growth of a Stokes field in the particular case of small-signal, steady-state scattering of a plane-wave pump field.

CHAPTER 2

RAMAN SCATTERING WITH A FOCUSSED PUMP BEAM

1. Introduction

As mentioned in the introduction to this thesis, under appropriate conditions, Stimulated Raman Scattering of a pump laser promotes the exponential growth of a Stokes beam with a rate that is proportional to the pump intensity. Often we may wish to maximise the conversion of energy from the pump to the Stokes frequency whilst the energy in the pump pulse, and the length of the Raman active medium are fixed. Clearly the intensity of the pump beam, and therefore the growth rate of the Stokes beam, are increased with a decrease in the pump beam diameter. We cannot conclude however, that the process of conversion is simply maximised simply by choosing an indefinitely small diameter for the pump; the diffraction of both the pump and Stokes beams are yet to be accounted for. Thus, in this chapter, we will consider the effects of pump focussing in Stimulated Raman Scattering. Our results will enable us to optimise the experimental conditions for an efficient conversion of energy and so determine the (minimum) pump energy required to attain some threshold. In addition, we will be able to predict the profile of the Stokes beam at the exit of the Raman active medium as a function of the focussing parameters.

In this Chapter, we start with a review of the earlier attempts that have been made to tackle the problem of pump focussing in Stimulated Raman Scattering. This is followed by the presentation of new results which are seen to encompass earlier approaches yet without their associated limitations. The approach used is that of a calculus of variations for a generalised Gaussian beam description of the Stokes field within the Lagrangian framework described in chapter 1.

2. Equations of motion for the Stokes field

In the following analysis we assume that the Stokes field growth is small signal, steady-state, and without competing processes. The conditions to be satisfied are respectively:

(a) The Stokes field is not large enough to deplete the pump or saturate the medium.
(b) The pump and Stokes field each have a bandwidth smaller than the Raman linewidth (Raymer et al, 1979).
(c) The gain and material dispersion of the medium favours the dominant growth of a field at the first Stokes frequency over higher order Raman processes (Perry et al, 1985).

Our starting point in the variational approach to the derivation of the Stokes field is the Lagrangian density for the electromagnetic field (see for example Goldstein, 1950):

$$\mathcal{L} = \frac{1}{2}\left[\underline{D}\cdot\underline{E} - \underline{B}\cdot\underline{H} \right] \tag{1}$$

and the Maxwell relation:

$$\frac{\partial \underline{H}}{\partial t} = \frac{1}{\mu_0}\, \underline{\nabla} \times \underline{E} \tag{2}$$

The pump and Stokes fields are defined as those components of the total field with frequencies ω_p and ω_s respectively. In the small signal regime, the pump field is unpertubed by the medium and its spatial distribution may therefore be regarded as given. Thus [1] and [2] apply to the field components at the Stokes frequency only, which we expand in the usual manner making explicit the rapidly varying part of the spatial variation in the z direction:

$$\underline{E} = \mathrm{Re}\left\{ \epsilon_s(\underline{r}) \exp\left[i\omega_s t - ik_s z \right] \underline{\hat{e}}_s \right\} \tag{3}$$

$$\underline{H} = \mathrm{Re}\left\{ h_s(\underline{r}) \exp\left[i\omega_s t - ik_s z \right] \underline{\hat{z}} \times \underline{\hat{e}}_s \right\} \tag{4}$$

$$k_s = \omega_s n_s / c \qquad [5]$$

where n_s is the refractive index at the Stokes frequency, $\epsilon_s(\underline{r})$ and $h_s(\underline{r})$ are slowly varying envelopes, and $\hat{e}_s$ defines the direction of the Stokes polarisation. In addition to (a), (b), and (c), it is assumed in this chapter that the pump field is a Gaussian beam and that the Stokes field is linearly polarised parallel to the pump field (for definiteness, this is taken to be in the x direction; see chapter 1, section 3). Then the fields $\underline{D}$, $\underline{B}$ can be written in terms of the electric and magnetic field vectors as follows:

$$\underline{B} = \mu_0 \underline{H} \;; \qquad \underline{D} = \varepsilon_0 \underline{E} + \underline{P} \qquad [6]$$

$$\underline{P} = \tfrac{3}{2}\, \varepsilon_0 |\epsilon_p|^2 \underline{\underline{\chi}}^{(R)} \underline{E} \qquad [7]$$

$$|\epsilon_p|^2 = |\epsilon_{p0}|^2 \left[\frac{W_{p0}}{W_p(z)}\right]^2 \exp\left[-\frac{2r^2}{W_p^2(z)}\right] \qquad [8]$$

$$W_p^2(z) = W_{p0}^2 \left[1 + 4\left(\frac{z-f}{k_p W_{p0}^2}\right)^2\right] \qquad [9]$$

where $\underline{\underline{\chi}}^{(R)}$ is the Raman susceptibility, the definition of which comes from Hanna et al (1979). Classically, the Stokes field $\underline{E}$ (with $\underline{H}$ given by [2]) will be that distribution for which the integral of the Lagrangian density is a minimum:

$$\delta \left\{ \iiiint \mathcal{L}(\underline{E}, \underline{H})\, dx\,dy\,dz\,dt \right\} = 0 \qquad [10]$$

We are justified in applying both the small divergence approximation and the paraxial approximation (see chapter 1) to the minimisation problem of [10]. This is because the contribution of the non-linear polarisation to the electric displacement in [6] is much less than that of the vacuum. Equally, we will find that the fractional deviation of the Stokes beam from a pure plane wave is negligible over a distance of λ_s, both radially and longitudinally. Again, appealing to chapter 1, we find that the x polarised Stokes field must therefore minimise the integral:

$$L = \frac{1}{4\mu_0\omega_s^2} \iiiint dx\,dy\,dz\,dt \left\{ \frac{3k_s^2 \chi_{xx}^{(R)}}{2n_s^2} |\epsilon_p \epsilon_s|^2 + 2\,\mathfrak{I}_m\left\{k_s \epsilon_s^* \frac{\partial \epsilon_s}{\partial z}\right\} - \left|\frac{\partial \epsilon_s}{\partial x}\right|^2 - \left|\frac{\partial \epsilon_s}{\partial y}\right|^2 \right\} \qquad [11]$$

whilst the Euler-Lagrange equation for the amplitude of the Stokes field in the above generates the paraxial ray equation:

$$\left\{ \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2i\kappa_s \frac{\partial}{\partial z} + \frac{3}{2}\frac{\kappa_s^2}{n_s^2} \chi^{(n)}_{xx} |\mathcal{E}_p|^2 \right\} \mathcal{E}_s = 0 \qquad [12]$$

Perry et al (1982, 1983) have posed the above as an eigenvector problem in the Hilbert space of Gauss-Laguerre functions which are the TEM freespace modes. The associated eigenvalues represent the growth of the Stokes beam on propagation through the gain medium. This decomposition of the problem relies on the separability of [12] for the particular 'refractive index' variation given by the Gaussian pump field (see the discussion in chapter 1, section 5). More recently, the same approach has been adopted by Gavrielides and Peterson (1986) to produce a numerical model valid in the large signal domain, i.e. taking into account pump depletion.

For the particular case $k_s = k_p$, Perry et al give their results for the variation of the three largest eigenvalues with the pump power. Although theirs is an exact (numerical) solution of [12], an approximate analytic treatment would in some cases be more desirable. For instance, one is generally interested in the component of the Stokes beam that couples into an optimally chosen TEM_{00} beam, whereas the spatial transverse profile of the Stokes beam at the exit of the gain medium is not readily recoverable from the Gauss-Laguerre eigenvectors.

The following treatment therefore models the Stokes field as a Gaussian beam throughout the medium, the parameters of which are chosen to minimise [11]. Our approximation consists of ignoring the coupling between this and higher order modes, although it will seen that this approach becomes exact either when the pump power is sufficiently large or sufficiently small. We therefore retain the Lagrange formulation, and substitute into [1] a Stokes field of the form:

$$\mathcal{E}_s(\underline{r}) = A(z) \exp\left[-i\, Q(z)\, r^2/2 \right] \qquad [13]$$

The amplitude A(z) and beam parameter Q(z) are now chosen so that [11] is a minimum. Thus we carry out the transverse integrations, and apply the Euler-Lagrange equations for the variation of $Q^*(z)$ and $A^*(z)$:

$$\frac{3\,k_s^2\,\chi_{xx}^{(R)}\,|\epsilon_{po}|^2 W_p^2(z)}{n_s^2\left[4 + iW_p^2(z)(Q-Q^*)\right]} = \frac{k_s Q' + Q^2}{[Q-Q^*]} \qquad [14]$$

$$k_s\frac{d\ln A}{dz} = \frac{|Q|^2 + k_s Q'}{[Q-Q^*]} + \frac{3\,k_s^2\,\chi_{xx}^{(R)}\,|\epsilon_{po}|^2 W_{po}^2[Q-Q^*]}{4\,n_s^2\left[4 + iW_p^2(z)(Q-Q^*)\right]} \qquad [15]$$

Equations [14] and [15] can be recast in terms of the normalised quantities as follows:

$$q^2 + \frac{\partial q}{\partial \zeta} + \frac{i\breve{P}_p}{2\kappa^2}\left[1+\zeta^2-(\kappa\,\Im_m q)^{-1}\right]^{-2} = 0 \qquad [16]$$

$$\frac{\partial \ln A}{\partial \zeta} = \frac{|q|^2 - q'}{q-q^*} + \frac{\breve{P}_p}{4\kappa}\left[1+\zeta^2-(\kappa\,\Im_m q)^{-1}\right]^{-1} \qquad [17]$$

where we have used the following definitions:

$$\breve{P}_p = \frac{3\kappa_s^2}{2n_s^2} W_{po}^2\,\chi_{xx}^{(R)}\,|\epsilon_{po}|^2 \qquad [18]$$

$$q = \frac{k_p W_{po}^2\,Q}{2\,k_s} \qquad [19]$$

$$\zeta = 2\left[\frac{z-f}{k_p W_{po}^2}\right] \qquad [20]$$

$$\kappa = k_s/k_p \qquad [21]$$

$\breve{P}_p$ is the 'normalised pump power' and ζ is the normalised longitudinal ordinate i.e. where possible, we have kept to the notation of Cotter et al (1975), whilst the definition of $\chi^{(R)}$ is that of Hanna et al (here assumed pure imaginary). Clearly if $\breve{P}_p = 0$, [16] and [17] reduce to the equations of motion for the spot-size, radius of curvature, and (complex) amplitude of a free-space Gaussian beam. When $\breve{P}_p \neq 0$ however, these equations can be used both to analyse the results of earlier authors in the domains of low and high pump power, *and* provide a more general description for the Stokes field for arbitrary $\breve{P}_p$; these are the respective goals of the sections which follow.

3. Solution to the equations of motion

3.1 Low pump power

We start by considering solutions to equations [16] and [17] in the limit of low pump power. We will first derive the general result for the Stokes amplitude and profile, and then show how this result can be applied to the design of a Raman gain cell.

If the normalised pump power $\tilde{P}_p$ is sufficiently small, the Stokes profile remains almost unchanged from its free space behaviour:

$$q^2 + \partial q / \partial \zeta = 0 \qquad [22]$$

In terms of normalised quantities, the solution of [22] is:

$$q = \frac{\mu \left[\mu(\zeta - \zeta_0) - i \right]}{\left[1 + \mu^2 (\zeta - \zeta_0)^2 \right]} \qquad [23]$$

where: $$\zeta_0 = \frac{2 f_s}{k_p W_{p0}^2} \qquad [24]$$

and: $$\mu = \frac{k_p W_{p0}^2}{k_s W_{s0}^2} \qquad [25]$$

Hence, ζ_0 is the distance of the Stokes focus from the pump focus in units of the pump confocal beam parameter, whilst μ is the ratio of pump to Stokes confocal beam parameter. This general case is depicted in figure 1 where the pump and Stokes beams have been enclosed by the gain medium. Of course, calculation of the Stokes field through equations [16] and [17] apply only to the field within the cell. Equally it is tacitly assumed that the finite transverse dimensions of the medium can be ignored.

With the free-space form for the Stokes profile, [17] can easily be solved to give the amplitude of the Stokes field at any point ζ in the gain medium:

$$A(\zeta) = A(\zeta_s)\frac{[1 - i\mu(\zeta_s - \zeta_0)]}{[1 - i\mu(\zeta - \zeta_0)]}\exp\left[\frac{\tilde{P}_p}{4\nu}\left[\tan^{-1}\frac{(\kappa+\mu^{-1})\zeta - \mu\zeta_0}{\nu} - \tan^{-1}\frac{(\kappa+\mu^{-1})\zeta_s - \mu\zeta_0}{\nu}\right]\right] \quad [26]$$

where: $$\nu = \left[1 + \kappa(\mu + \mu^{-1}) + \kappa^2 + \mu\kappa\zeta_0^2\right]^{1/2} \quad [27]$$

and $A(\zeta_s)$ is the Stokes amplitude at the entrance to the medium. The total power in the Stokes beam can be evaluated from the above (using [23]):

$$P_s(\zeta) = P_s(\zeta_s)\exp\left[\frac{\tilde{P}_p}{2\nu}\left[\tan^{-1}\frac{(\kappa+\mu^{-1})\zeta - \mu\zeta_0}{\nu} - \tan^{-1}\frac{(\kappa+\mu^{-1})\zeta_s - \mu\zeta_0}{\nu}\right]\right] \quad [28]$$

The justification for using the free-space profile [23] in deriving [28] is that our result for the Stokes power is then directly comparable with those of earlier workers. In fact Boyd et al (1969) have obtained exactly the same result using an 'overlap integral' method. Whilst [28] is also related to the result obtained by Christov and Tomov (1985). Also, allowing for typographical errors, the same result has been obtained by Trutna and Byer (1980). Examination of [16] reveals however, that to first order in $\tilde{P}_p$, the third term also contributes to the gain as described by [17]. Thus we find that even for low pump powers, the effect of the pump power on the Stokes beam profile can be significant. However, this component can be shown to be identically zero for the particular initial Stokes profile satisfying:

$$\zeta_0 = 0; \quad \mu = 1$$

which is just that the pump and Stokes beams share a confocal plane, and have equal confocal parameters. These are the conditions are chosen by Trutna and Byer to maximise their expression, based on [28], for the Stokes gain.

Strictly speaking, Trutna and Byer obtained a maximum gain through optimal choice of the confocal parameters belonging to both the pump and the Stokes field under the assumption that the beams share a confocal plane. They rightly concluded that the Stokes gain would be a maximum - in the limit of tight focussing for the pump - if the confocal parameters were equal. Thus in a cavity designed to give rise to a self-reproducing pump beam, the optimal choice of confocal parameters is also that which gives rise to a self-reproducing Stokes beam. We note in passing that for a cavity design other than that of Trutna and Byer wherein the pump beam is not tightly focussed, the condition $\mu=1$ does not maximise the Stokes gain. In this case the characteristics and growth rate of the Stokes beam will be a result of the (competing) tendencies towards a beam that is self-reproducing, and one that has maximum gain.

Within the variational framework of this chapter however, the condition above is a *necessary prerequisite* for the validity of [28]. Therefore we will proceed assuming that these conditions are met by the design of the Raman amplifier, so that by virtue of our more general approach, we will then be in a position to determine the validity of the low gain approximation. In this case, the power gain for the Stokes beam is found from [28] to be:

$$P_s(\zeta) = P_s(\zeta_s) \exp\left[\frac{\tilde{P}_p\, \Theta(\zeta;\zeta_s)}{2(1+\kappa)} \right] \qquad [29]$$

where $\theta(\zeta;\zeta_s)$ is the (dimensionless) parameter:

$$\Theta(\zeta;\zeta_s) = \tan^{-1}\zeta - \tan^{-1}\zeta_s \qquad [30]$$

If we now compare the magnitude of the discarded term in [16] with those used to define the free-space profile [23], then we find that the low gain approximation is consistent with the requirement:

$$\hat{P}_p \ll 2(1+\kappa)^2(1+\zeta^2) \qquad [31a]$$

which must therefore be regarded as a necessary condition for the validity of [29]. In this form, the constraint above is rather unsatisfactory since it depends strongly on the length of the Raman gain medium. However, a more accurate constraint can be found from comparing the result [29], with an exact solution for the Stokes exponential gain which we will anticipate from the results of section 3.3. Thus by expanding the exponential gain in increasing powers of the normalised pump power, we find it is necessary that:

$$\hat{P}_p \ll \left[32(1+\kappa)^5\right]^{1/2} \qquad [31b]$$

whence in the low pump power domain, [29] gives an accurate measure of the Stokes exponential gain.

3.2 High pump power

As the pump-power is increased, the Stokes profile will deviate from the free-space form given by [23]. Hence the extent of the profile is governed by the competing effects of diffraction and gain-focussing determined respectively by the first and third terms of equation [16]. When the pump power is sufficiently high, effect of the gain-focussing is to confine the Stokes spot-size to an area well within that of the 'guiding' pump, i.e. in the limit of high pump power we expect:

$$W_p^2(z) >> W_s^2(z) \qquad [32]$$

The Stokes spot-size can be defined (using [13] and [19], in terms of the normalised variable q; whilst the pump spot-size can be defined (using [9] and [20] in terms of the normalised co-ordinate ζ. Thus we may rewrite [32] as:

$$1+\zeta^2 >> -\left[\kappa \, \mathrm{Im}\, q\right]^{-1}. \qquad [33]$$

(*If* a TEM_{00} Stokes mode exists, then the imaginary part of q must always be negetive.) Using [33], [16] becomes:

$$q^2 + \frac{\partial q}{\partial \zeta} + \frac{i\tilde{P}_r}{2\kappa^2(1+\zeta^2)^2} = 0 \qquad [34]$$

We note that the same result can be obtained by retaining only the zeroth and quadratic terms in the expansion of $|\epsilon_p|^2$ in powers of r in equation [8]. Hence this approach is just that of the parabolic-index profile approximation considered by Cotter et al (1975). In this chapter however, we proceed to solve for q without the additional approximations made in that work.

Equation [34] is a Ricatti equation, and can be cast as a linear second order differential equation by making the usual change of variable:

$$q = \frac{1}{v}\frac{dv}{d\zeta} \quad ; \quad \gamma = \left[1 + \frac{i\tilde{P}_r}{2\kappa^2}\right]^{1/2} \qquad [35]$$

whence: $$\frac{d^2 V}{d\zeta^2} + \frac{(\gamma^2-1)\,V}{(1+\zeta^2)^2} = 0 \qquad [36]$$

By substitution or otherwise, the solution of [36] can be shown to be:

$$V(\zeta) = V_0\left[1+\zeta^2\right]^{1/2} \cos\left[\gamma\,\Theta(\zeta;\zeta_s) + \phi\right] \qquad [37]$$

and V_0, ϕ are (complex) arbitrary constants. From the substitution in [35], the complex parameter q can be recovered:

$$q(\zeta) = \frac{\zeta - \gamma\tan(\gamma\Theta + \phi)}{1+\zeta^2} \qquad [38]$$

where now ϕ can be interpreted interms of the Stokes parameters at the entrance to the gain medium:

$$\phi = \tan^{-1}\left[\frac{\zeta_s - (1+\zeta_s^2)\,q(\zeta_s)}{\gamma}\right] \qquad [39]$$

The equation for the amplitude of the Stokes field can now be derived from [16] and [17] and making use of [33]:

$$\frac{d\,\mathrm{Ln}\,A}{d\zeta} = -q + \frac{\tilde{P}_p}{4\kappa(1+\zeta^2)} \qquad [40]$$

Again, recalling the substitution in [35] and the result for $V(\zeta)$ in [37], the amplitude can be written down without further calculation:

$$A(\zeta) = A(\zeta_s)\left[\frac{1+\zeta_s^2}{1+\zeta^2}\right]^{1/2} \frac{\cos(\phi)}{\cos(\gamma\Theta+\phi)} \exp\left[\frac{\tilde{P}_p\,\Theta}{4\kappa}\right] \qquad [41]$$

The condition [33] can now be stated using [38] as:

$$\mathrm{Im}\left\{\kappa\gamma\tan(\gamma\Theta+\phi)\right\} >> 1 \qquad [42]$$

Equations [38] and [41], in conjunction with [42], describe the behaviour of the Stokes field under the quadratic index profile approximation.

Just as for the case of low gain, we proceed with an example maing use of these results. In particular, we consider the initiation of the stimulated process from spontaneous scattering at $\zeta = \zeta_s$. We will take the initial field to be a Gaussian beam with zero spot-size and zero radius of curvature, whence from [39]:

$$q(\zeta_s) = \infty - i\infty \tag{43}$$

After a short distance into the gain medium, the complex parameter q from [38] obeys:

$$q(\zeta) \simeq \frac{\zeta - i\gamma}{1 + \zeta^2} \tag{44}$$

and the Stokes amplitude is:

$$A(\zeta) \simeq 2iA(\zeta_s)\left[\frac{1+\zeta_s^2}{1+\zeta^2}\right]^{1/2} \exp\left[\left(\frac{\tilde{\rho}_f}{4\kappa} + i\gamma\right)\Theta(\zeta;\zeta_s)\right] \tag{45}$$

The condition for the parabolic index profile approximation is now:

$$\kappa\, \mathfrak{I}_m(\gamma) >> 1 \tag{46}$$

and in deriving [44] and [45], use has been made of the additional constraint:

$$\left| \exp\left[-2i\gamma\Theta(\zeta;\zeta_s)\right] \right| >> 1 \; ; \quad \zeta_s \lessapprox \zeta \lessapprox \zeta_e \tag{47}$$

This is a simplifying assumption designed to ensure that the cosine terms in [41] effectively collapse into the dominant exponential component. The value of ζ for which [47] becomes true depends on the magnitude of the gain: the higher the gain, the earlier will this constraint be satisfied and therefore will q approach the the particular form [44].

Defining the real and imaginary parts of Q in terms of the spot-size and radius of curvature (see for instance Yariv, 1975):

$$Q = \frac{k_s}{R_s(z)} - \frac{2i}{W_s^2(z)} \tag{48}$$

then we find that [44] implies that the Stokes beam has a radius of curvature:

$$R_s = \frac{k_p W_{po}^2}{2} \left[\frac{1+\zeta^2}{\zeta + \mathcal{I}m(\gamma)} \right] \qquad [49]$$

and spot-size given by:

$$W_s^2 = W_p^2(z) \,/\, [\, \kappa\, \mathcal{R}e(\gamma)\,] \qquad [50]$$

Hence the Stokes field is a Gaussian beam with propagation characteristics similar to that of a free-space beam, but with a distorted phase front, and a spot-size that is everywhere narrower than its free-space equivalent.

Equations [44] and [45] describe the 'matched mode' behaviour of the Stokes field in that the complex parameter $q(\zeta)$ and amplitude $A(\zeta)$ have become independent of the initial parameter $q(\zeta_s)$. This is a generalisation of a concept first introduced in this context by Cotter et al. The magnitude of the pump power, through the left hand side of [47], is seen to determine how quickly the initial Stokes profile tends towards the matched mode profile. In fact, if instead of [43], the initial parameter is made to satisfy the matched mode condition at $\zeta = \zeta_s$:

$$q(\zeta_s) = \frac{\zeta_s - i\gamma}{1+\zeta_s^2} \qquad [51]$$

then the $q(\zeta)$ remains unchanged from its matched mode value throughout the medium.

These results can be compared with those of Cotter et al by taking the limit for high pump power of the complex parameter given by [34]. Under these conditions, the Stokes power is:

$$P_s(\zeta) \simeq 4\, P_s(\zeta_s) \exp\left[\frac{\breve{P}_p - 2\sqrt{\breve{P}_p}}{2\kappa}\, \Theta(\zeta;\zeta_s) \right] \qquad [52]$$

where now [46] becomes:

$$\breve{P}_p \gg 4 \qquad [53]$$

Therefore, the results of Cotter et al represent the high pump power limit of the matched mode solution. Note that by virtue of [53], the expression for the Stokes power in [52] is valid only when the net exponential gain is greater than zero. Thus the explanation based on this result which was advanced by Cotter for the behaviour of the Stokes beam at low pump power is spurious. Note also that [52] in conjunction with [53], describes a Stokes power similar to that obtained from the low pump power calculation of the previous section. The first term in the exponent is greater by a factor $(1 + k)/\kappa$, whilst the additional second term represents a reduction in gain due to the increased diffraction of the Stokes field in the presence of gain-focussing.

3.3 Matched mode

Following the discussion in the previous section, we seek an *exact* matched mode solution to the equations of motion [16] and [17] without making the parabolic index profile approximation. The result will then be an analytic description for the Stokes field that will simultaneously cope with the low gain conditions as for example in a multipass Raman gain cell, and the high gain conditions encountered in a single pass Raman generator. In either case, the matched mode condition may be arrived at through one of two routes:

(a) An initially unmatched mode perturbed by the gain medium to a point where the spot-size and radius of curvature have converged upon that of the matched mode. From the previous section, we find this condition will generally be satisfied if:

$$\left| \exp\left[-2i\gamma\,\theta(\zeta;\zeta_s) \right] \right| \gg 1 \qquad [47]$$

(b) An injected field which is a Gaussian beam with spot-size and radius chosen to satisfy the matched mode condition at $\zeta = \zeta_s$.

The matched mode solution to [16] may be derived from a substitution of the form:

$$q = \frac{\alpha - i\beta + \epsilon\zeta}{1 + \zeta^2} \qquad [54]$$

where α, β, and ϵ are real, and $\beta > 0$. Upon equating equal powers of ζ we obtain:

$$\epsilon = 1 \qquad [55]$$

$$\alpha = \sqrt{\beta^2 - 1} \qquad [56]$$

$$\beta\,\tilde{\rho}_r = 4\left[1 + \kappa\beta\right]^2 \sqrt{\beta^2 - 1} \qquad [57]$$

whilst the amplitude now satisfies:

$$\frac{d\,\mathrm{Ln}\,A}{d\zeta} = \frac{1}{[1+\zeta^2]}\left[\frac{\breve{P}_p\,\beta}{4(1+\kappa\beta)} + i\left(\beta+\frac{1}{\beta}\right)\right] - \frac{\zeta}{[1+\zeta^2]} \qquad [58]$$

and so the Stokes power is easily found to be:

$$P_S(\zeta) = P_S(\zeta_s)\exp\left[G(\breve{P}_p,\kappa;\theta)\right] \qquad [59]$$

where $G(\breve{P}_p,k)$ is the matched mode exponential power gain:

$$G(\breve{P}_p,\kappa;\theta) = \frac{\breve{P}_p\,\beta\,\theta}{2(1+\kappa\beta)} \qquad [60]$$

and the matched mode complex parameter q is:

$$q = \frac{\sqrt{\beta^2-1} - i\beta + \zeta}{[1+\zeta^2]} \qquad [61]$$

and β is given by the solution of [57]. That [59] and [61] encompass the high and low gain results of the previous sections can readily be seen from the limiting values for β:

$\breve{P}_p$ small:
$$\beta = 1 + \frac{\breve{P}_p^2}{32(1+\kappa)^5} + \mathcal{O}(\breve{P}_p^3)$$
$$\Rightarrow \quad G = \theta\left[\frac{\breve{P}_p}{2(1+\kappa)} + \frac{\breve{P}_p^3}{64(1+\kappa)^6} + \mathcal{O}(\breve{P}_p^4)\right] \qquad [62]$$

$\breve{P}_p$ large:
$$\beta = \frac{\breve{P}_p^{1/2}}{2\kappa} - \frac{1}{\kappa} + \mathcal{O}(\breve{P}_p^{-1/2})$$
$$\Rightarrow \quad G = \theta\left[\frac{\breve{P}_p}{2\kappa} - \frac{\breve{P}_p^{1/2}}{\kappa} + \mathcal{O}(\breve{P}_p^{-1/2})\right] \qquad [63]$$

Note that comparison of the first and second terms in the expansions for the exponential gain, confirms the constraints for the validity of each of the results in the domains of low and high pump power in the previous sections (equations [31b] and [53] respectively).

We note in passing that the gain-focussed Stokes beam becomes ever more confined with increasing pump power and therefore can expect the parabolic index profile approximation discussed in section 3.2 to give increasingly accurate results. Thus the coupling between modes will eventually vanish and the high gain limit given by [59-61] and [63] will give the exact solution to [12]. Further it is recalled that the result first obtained by

Trutna and Byer is effectively that of a first order perturbation theory (in the pump intensity) applied to a Stokes field expansion in free-space TEM_{nm} modes. Hence for sufficiently low pump powers, the low gain limit given by [62] will also give an exact solution to [12].

For the general matched mode result, it may be of interest to know the spot-size and radius of curvature at any point ζ in the medium. Comparison of [54] with [44] reveals that the substitutions:

$$\mathrm{Re}\ \{\ \gamma\ \} \rightarrow \beta \qquad [64]$$

$$\mathrm{Im}\ \{\ \gamma\ \} \rightarrow \alpha = \sqrt{\beta^2 - 1} \qquad [65]$$

into [49] and [50] gives the general results:

$$R_s = \frac{K_p W_{p0}^2}{2} \cdot \frac{1 + \zeta^2}{\zeta + \sqrt{\beta^2 - 1}} \qquad [66]$$

$$W_s^2 = \frac{W_p^2(z)}{\kappa\ \beta} \qquad [67]$$

where again β is given by the solution of [57]. The radius and spot-size at the end of the gain medium can be found simply by substituting $\zeta = \zeta_e$ into [66] and [67] respectively. It is clear from these results that the radius of curvature and the spot-size are smaller than that of the equivalent free-space mode which has $\mu = 1$ and shares a focal plane with the pump beam.

4. Discussion

We now compare the results predicted by [60], [61] with both the approximate results of sections 3.1 and 3.2, and the numerical results of Perry et al. In the following, we will assume that the pump focussing conditions are such that $\theta(\zeta;\zeta_s) \simeq \pi$, and therefore that the Stokes exponential power gain is given by:

$$G(\breve{P}_p, \kappa) = \frac{\breve{P}_p \beta \pi}{2(1+\kappa\beta)} \qquad [68]$$

The equation [57] has been solved for β numerically, and a plot of β versus $\breve{P}_p$ for various values of κ is given in figure 2. These results can be used to find the matched mode exponential gain $G(\breve{P}_p,\kappa)$ in [60], and the spot-size and radius of curvature in [66] and [67]. In figure 3 we compare the matched mode gain with the gain predicted by [62] and [63]. As expected, it is seen that the limiting cases are satisfactorily modelled as $\breve{P}_p \to 0$ and $\breve{P}_p \to \infty$ respectively. For the chosen value of $\kappa = 1$, we observe that the predictions of the high and low pump power approximations are equal at $\breve{P}_p = 16$ (the high pump power solution thereafter being closer than the low pump power solution to the matched mode gain). In this aspect then, this is the point at which the conventional models are least satisfactory; there being about 15% deviation from the matched mode gain.

It is also of interest to compare these results with those obtained by Perry et al (1982) (see figure 1 of that work). First it is necessary to make explicit the connection between the symbols used in their work, and those adopted in this chapter. Table 1 provides a summary of the pertinent relationships:

Table 1

Description	Perry et al	This chapter
Dimensionless parameter	μ	$\dfrac{\kappa}{1+\kappa}$
Gain coefficient	G_p	$\hat{P}_p / 4$
Real part of eigenvalue	$Re\{\lambda\}$	$\dfrac{\hat{P}_p \beta}{4(1+\kappa\beta)}$
Normalised gain	$Re\{\lambda\} / G_p$	$\dfrac{\beta}{1+\kappa\beta}$

In figure 4 we use these relationships to compare the matched mode gain with the gains predicted by Perry et al (at $\kappa = 1$) for the first and second (rotationally symmetric) eigenfunctions. Clearly the matched mode gain is consistently close to the gain of the first eigenfunction, and the good correspondence between these results therefore lends support to the analytic model and the results [60], [61] and [57]. Encouraged by this comparison, we present in figure 5 the matched mode gain for various values of κ found by applying the numerical solution of [57] to [60].

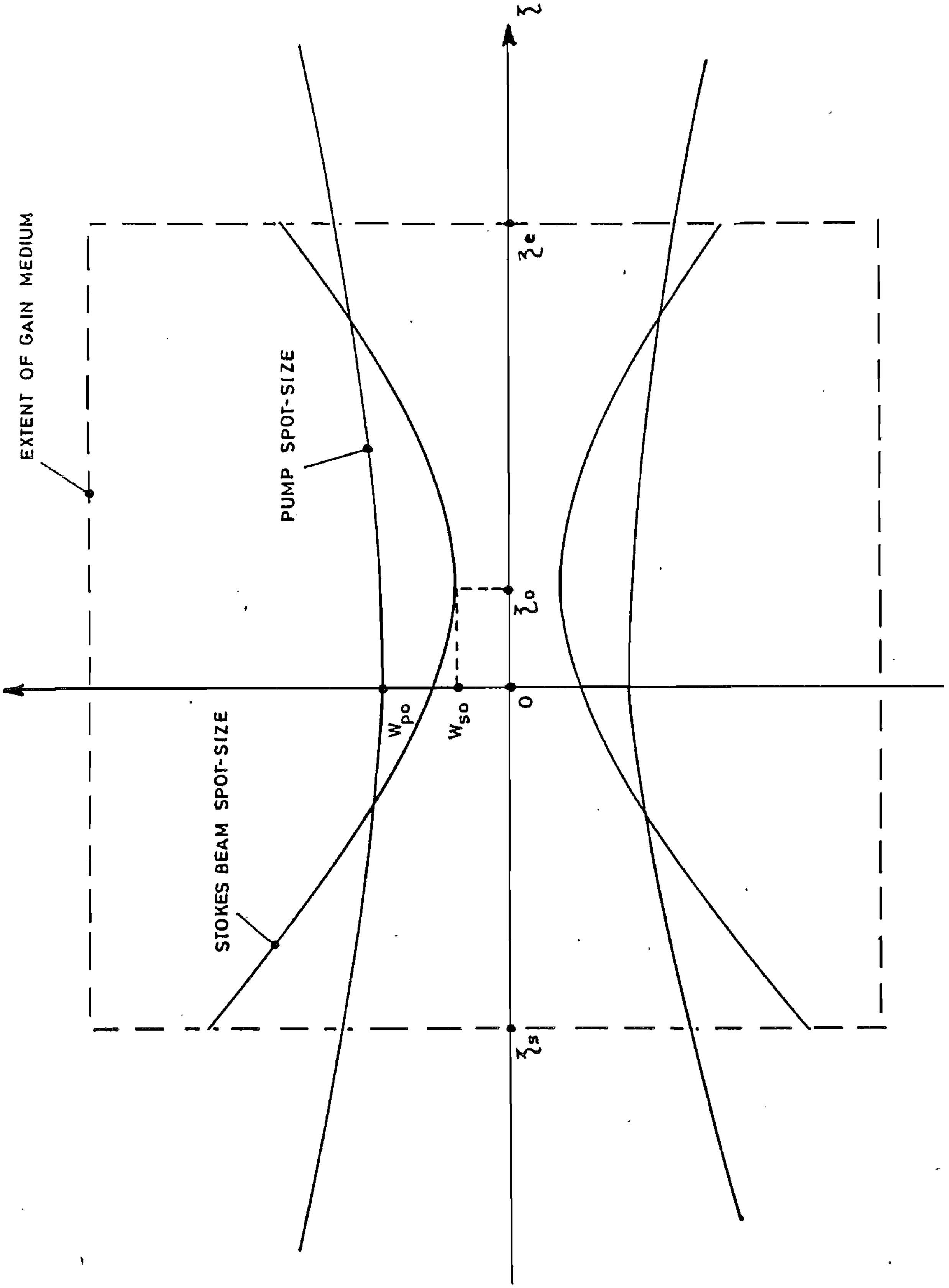
It is possible to further test the accuracy of our supposition that the lowest order mode is essentially a Gaussian beam by comparing the matched mode profile with that predicted by Perry et al. With reference to figure 6, we have used the normalised coordinate $r/w_*(z) = r\sqrt{\kappa\beta}/w_p(z)$, and find that once again, at least for the values $\hat{P}_p = 40$ and $\kappa = 1$, there is good agreement between the results.

5. Conclusions

In this chapter we have presented a novel analytic model for the evolution of a Stokes field in a Raman active medium excited by a focussed pump beam. We have shown that the results of this model are valid for a wide range of pump powers, and that in the limits of high and low pump power, they reproduce the results of earlier workers. We have therefore been able to identify constraints which in this context define the domains of high and low pump power. Excellent agreement has been obtained in comparison with an exact numerical treatment.

<u>FIGURE 1</u>

Configuration of pump and Stokes beam in a gain medium.



<u>FIGURE 2</u>

Dimensionless parameter β versus normalised pump power for various κ.

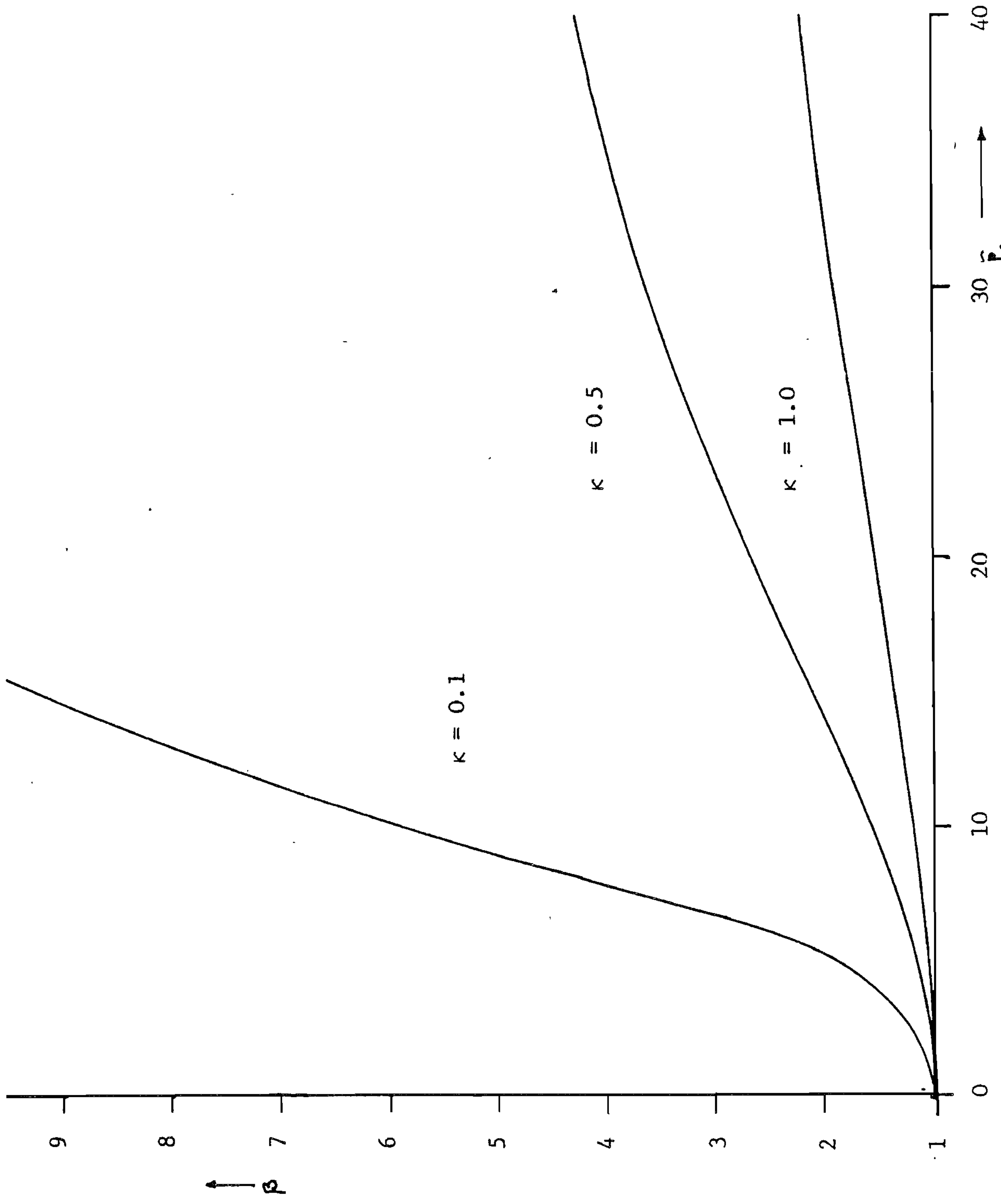


FIGURE 3

tokes exponential gain as predicted by matched-mode, low gain, and igh gain theory.

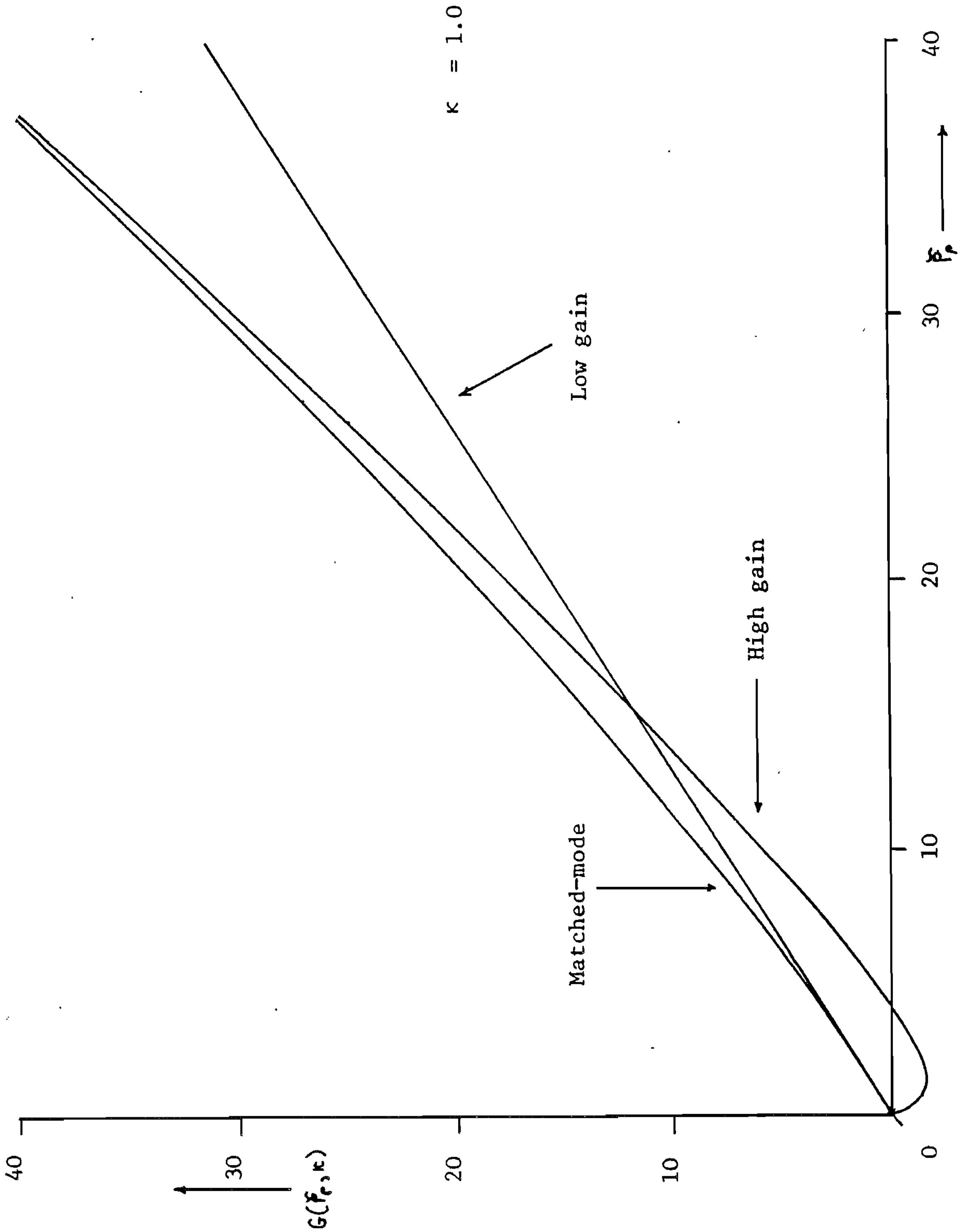


FIGURE 4

Stokes exponential gain as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.

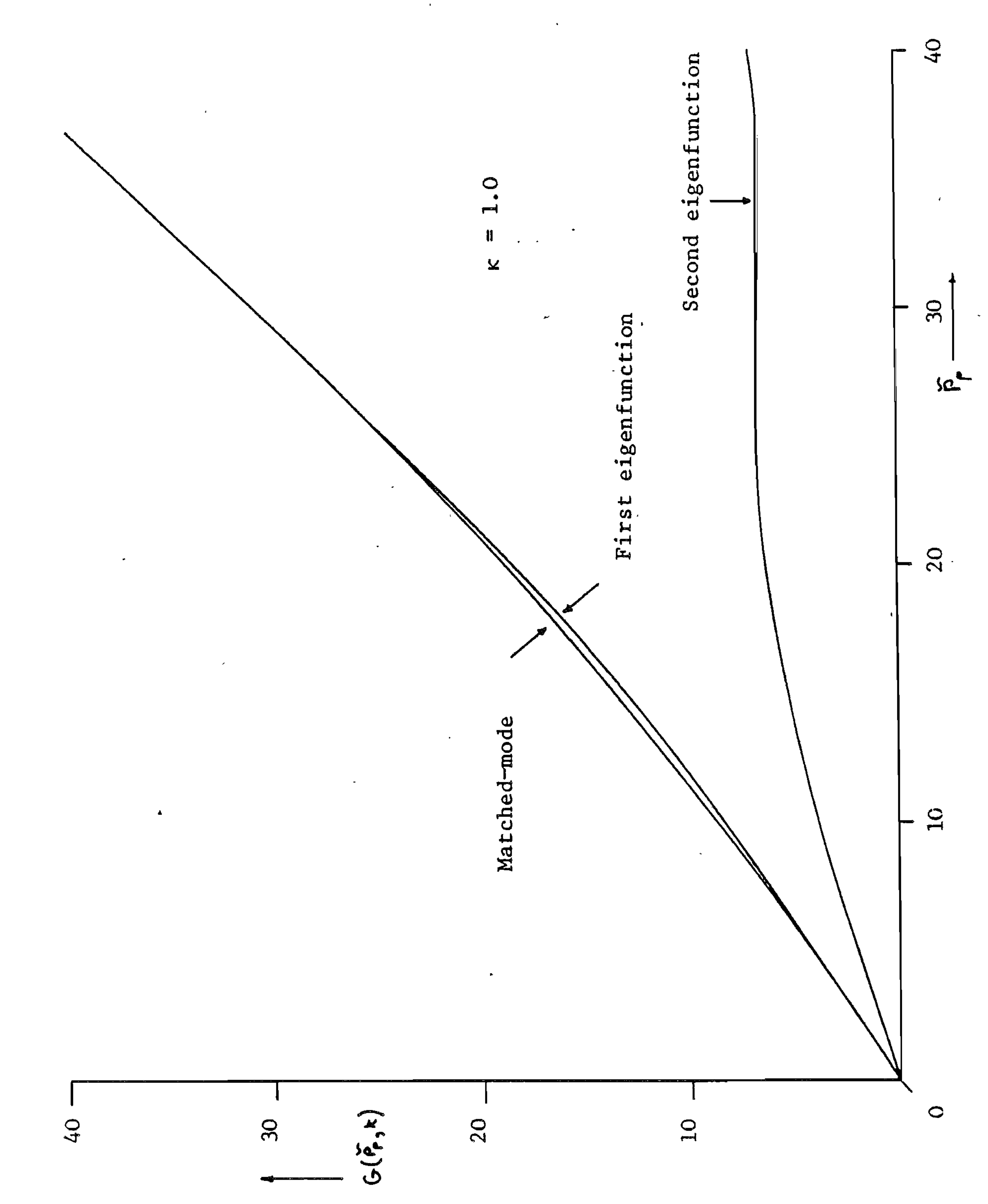


FIGURE 5

Stokes exponential gain as predicted by the matched-mode theory versus normalised pump power for various κ.

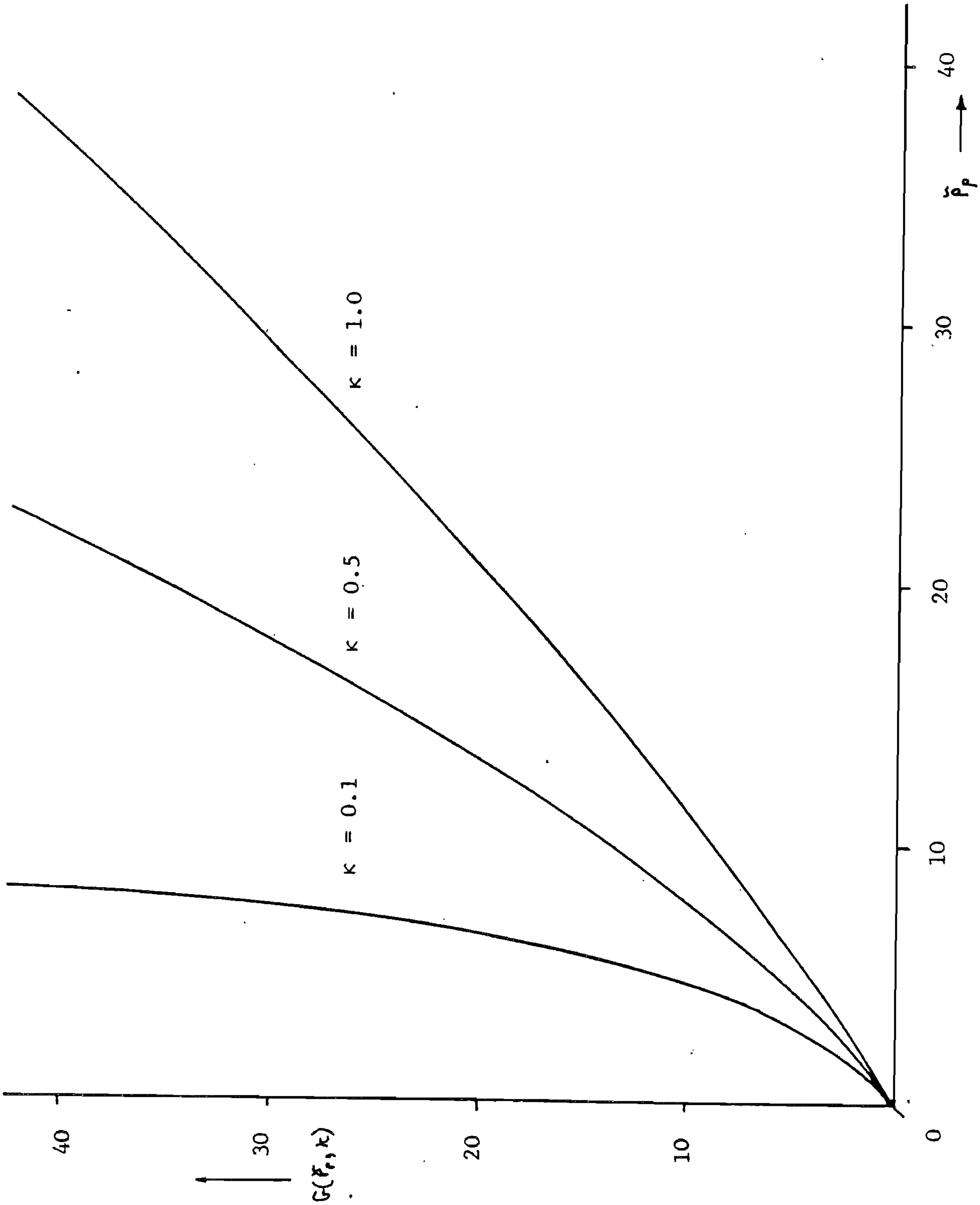
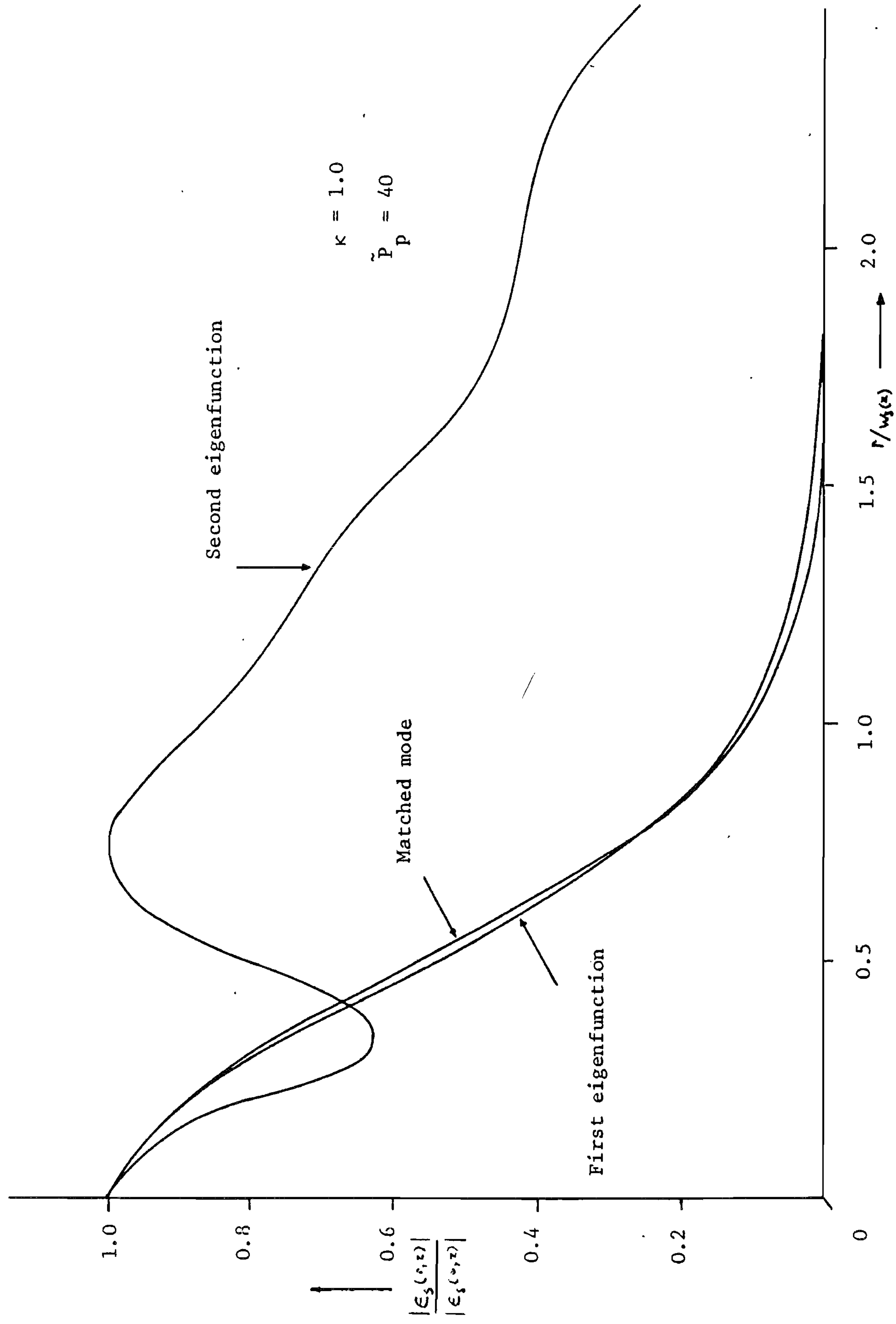


FIGURE 6

Stokes profile as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.



CHAPTER 3

RAMAN SCATTERING IN A WAVEGUIDE

1. Introduction

In chapter 2 we investigated the effects of using a focussed pump beam to drive the Raman active medium. In this chapter the same motivation, namely that of a reduced threshold for the pump power, will encourage us to consider the effects of Raman generation in a waveguide. This time the spatial confinement of both pump and Stokes fields is achieved as each suffers multiple reflections on its journey down the guide. Once again the aim is to characterise the behaviour of the Stokes field at the output of the system in terms of the experimentally adjustable parameters, which in this case will include the diameter and length of the waveguide as well as the pump power and focussing conditions.

Raman generation in a waveguide has a potential advantage over generation in free-space in that the 'interaction length' between pump and Stokes fields can be much greater than would otherwise be possible. The process of tight focussing, which may be necessary to achieve the required pump intensities, enhances the diffraction of both pump and Stokes beams, so that the (effective) interaction length remains always of the order of a reciprocal confocal beam parameter (see chapter 2).

In this chapter we will concentrate on the application of hollow dielectric waveguides (capillaries) to Raman Scattering in gases, and thus hope to produce some results relevant to the experimental work at Southampton University (see for instance Berry and Hanna, 1982, 1983). The first reports of the use of capillaries in Raman Scattering were by Rabinowitz et al (1976) and Hartig and Schmidt (1979), the latter reporting greatly enhanced efficiency through a reduction in pump threshold power. Theoretical analysis of Raman Scattering in (non-metallic) waveguides has been attempted by Yeung and Yariv (1978) and more recently by Urquhart and Laybourn (1986). Although these two references deal with approximate techniques for the generation of Stokes fields in glass fibres, the simplified numerical model of Urquhart and Laybourn will be shown, under most circumstances, to be applicable also to the generation of Raman Scattering in capillaries (this is possible because terms which they employ to describe the wavelength dependent material absorbtion within the fibre may alternatively be used to describe the non-equal losses suffered by the pump and Stokes fields on reflection at the waveguide walls). However, unlike Urquhart and Laybourn, we will be concerned with the stimulated scattering process in the small signal regime so that depletion of the pump field can be ignored.

In section 2 we present an approximate model for the propagation of a field in a glass capillary and in section 3 we show how this theory can be applied to the description of both the pump and the Stokes fields. We conclude in section 4 with a discussion on the choice of optimum guide characteristics for a variety of experimental scenarios.

2. Electromagnetic theory

2.1 Solutions to the wave equation

In this section we will make use of both the classical mode theory of the dielectric waveguide (see for example Marcatili and Schmeltzer, 1964) and the Lagrangian formulation of the general inhomogeneous problem. We will borrow from the results of chapter 1 to obtain a Lagrangian density incorporating the non-linear Raman polarisation which will then be solved by the method of calculus of variations. First however we must discuss the behaviour of electromagnetic fields in an empty capillary in order that we may understand the behaviour of the pump field and also be in a position to later suggest suitable trial functions for the approximate (variational) Stokes field.

Figure 1 depicts the dielectric waveguide referred to in this chapter. The axis of the guide is made to lie along the z axis, and the core region has a radius 'a', whilst the cladding is supposed large enough to be considered as infinite. The classical modes of a dielectric guide are found from solving the Maxwell equations in a cylindrical polar co-ordinate system. For consistency, we will briefly show how these modes can be derived from the Lagrangain density of chapter 1 where now there is neither refractive index variation, nor nonlinear polarisation of the medium. The Euler-Lagrange equations in such a case are easily found to be:

$$\Box \underline{A} = 0 \; ; \qquad \underline{\nabla}.\underline{A} = 0 \qquad [1]$$

and we recall that $\underline{A}$ is related to the electric and magnetic fields thus:

$$\underline{B} = \underline{\nabla} \times \underline{A} \qquad [2]$$

$$\underline{E} = -\delta \underline{A}/\delta t \qquad [3]$$

By simple manipulations, it is easy to show that $\underline{B}$ (and therefore $\underline{H}$) and $\underline{E}$ also satisfy the wave equation:

$$\Box \underline{H} = 0 \; ; \qquad \underline{\nabla} . \underline{H} = 0 \qquad [4]$$

$$\Box \underline{E} = 0 \; ; \qquad \underline{\nabla} . \underline{E} = 0 \qquad [5]$$

and now: $$\underline{\nabla} \times \underline{E} = -\mu_o \delta \underline{H} / \delta t \qquad [6]$$

It follows from the above that just two of the six components of the electromagnetic field are independent. Traditionally, these are taken to be the longitudinal fields E_z and H_z. Before we write the remaining fields in terms of these two, it is convenient to remove the Fourier components describing the z, t and θ variation of all the fields:

$$(\underline{E}, \underline{H}) \sim \exp(i\omega t - i\beta z + i\nu\theta) \qquad [7]$$

The sign of β is fixed indicating a forward propagating wave, whilst ν can take either sign. Making use of the above and relations [4-6], it is now possible to express (in a closed form) the remaining fields in terms of the longitudinal components:

$$E_r = -\frac{a^2}{U^2}\left[\frac{\nu\omega\mu_o}{r} H_z + i\beta \frac{\partial E_z}{\partial r}\right] \qquad [8]$$

$$E_\theta = -\frac{a^2}{U^2}\left[i\omega\mu_o \frac{\partial H_z}{\partial r} - \frac{\beta\nu}{r} E_z\right] \qquad [9]$$

$$H_r = \frac{a^2}{U^2}\left[\frac{\nu n_1^2 \varepsilon_o \omega E_z}{r} - i\beta \frac{\partial H_z}{\partial r}\right] \qquad [10]$$

$$H_\theta = \frac{a^2}{U^2}\left[i n_1^2 \varepsilon_o \omega \frac{\partial E_z}{\partial r} + \frac{\nu\beta}{r} H_z\right] \qquad [11]$$

where E_z and H_z are found from:

$$\left[\frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r} - \frac{\nu^2}{r^2} + \frac{U^2}{a^2}\right]\left(E_z, H_z\right) = 0 \qquad [12]$$

In this chapter the wavenumber k is just $2\pi/\lambda$, the refractive index at both the pump and Stokes frequencies is taken to be n_1 in the core region, and n_2 in the cladding.

Equations [12] admit solutions of the form:

$$(E_z, H_z) \sim J_\nu(ur/a) \text{ and } K_\nu(ur/a) \qquad [13]$$

i.e. Bessel functions of the first and second kinds. Here, u is a dimensionless parameter given by:

$$u = a[n_1{}^2k^2 - \beta^2]^{1/2} \qquad [14]$$

The appropriate solutions must be chosen with due consideration given to the boundary conditions on each of the field components. In practice, it is usual to solve the propagation problems in the core and cladding regions independently, and then derive the eigenmodes from applying the continuity conditions for the fields at the waveguide walls (i.e. the core/cladding interface). In our case, we have $n_2 > n_1$ so that a mode of the core also has an oscillating component in the cladding - i.e. the 'core modes' are not properly confined by the guide. However, by insisting that the fields vanish at some large (but otherwise arbitrary) radial distance from the guide axis, we ensure that only one of the two types of solution is permitted in the cladding. This in turn creates the desired eigenvalue problem by virtue of the boundary conditions on the fields at the core-cladding interface. The problem is thereby considerably simplified without compromising the basic results.

The appropriate solution for the longitudinal field components in the core region is the non-singular Bessel function of the first kind. In the cladding region, following the discussion above, we retain only the Bessel function of the second kind:

$$\{E_z, H_z\} \sim \begin{cases} J_\nu(ur/a) & \text{:core} \\ K_\nu(wr/a) & \text{:cladding} \end{cases} \qquad [15]$$

where w is a dimensionless parameter given by:

$$w = a[\beta^2 - n_2{}^2k^2]^{1/2} \qquad [16]$$

The definition of w is borrowed from the theory of propagation in fibres where usually $n_2 < n_1$; in our case w is imaginary.

Combining [7] and [15] gives two distinct solutions which may be substituted into the relations [8-11]:

$$\{E_z, H_z\} = \{A, B\} \exp\left[i\omega t - i\beta z + i\nu\theta\right] \begin{cases} J_{|\nu|}(ur/a) \big/ J_{|\nu|}(u) & \text{:core} \\ K_{|\nu|}(wr/a) \big/ K_{|\nu|}(w) & \text{:cladding} \end{cases} \quad [17]$$

where A and B are constants.

2.2 Boundary conditions

It can be seen from our choice of normalisation that both E_z and H_z are continuous at r=a (for all θ and z). In fact the boundary conditions on the electromagnetic fields in a dielectric waveguide are that the tangential components of both fields are continuous at the waveguide walls. Thus it remains only to ensure that E_θ and H_θ are continuous at r=a. Substitution of [17] into [9] and [11] and application of the continuity condition gives:

$$\frac{A}{B} = \frac{-i\omega\mu_0}{\beta\nu}\,\frac{u^2w^2}{u^2+w^2}\left[\frac{J_\nu'(u)}{uJ_\nu(u)} + \frac{K_\nu'(w)}{wK_\nu(w)}\right] = \frac{-i\nu\beta}{\omega\varepsilon_0}\cdot\frac{u^2+w^2}{u^2w^2}\left[\frac{n_1^2J_\nu'(u)}{uJ_\nu(u)} + n_2^2\frac{K_\nu'(w)}{wK_\nu(w)}\right]^{-1} \quad [18]$$

The above implies both a relationship between the coeffiecients A and B, and a constraint on the possible values for β through u and w i.e. the above determines the eigenvalues for the propagation problem in a dielectric waveguide.

Modes propagating in a capillary waveguide suffer losses on reflection at the waveguide walls. It will be seen that the extent of these losses depends on the radius of the core region and the wavelength of the propagating radiation: the losses are low when the ratio λ/a is small. Generally we may assume that the capillary has been designed so that losses are small and will therefore be justified in assuming:

$$ka \gg 1 \quad [19]$$

For sufficiently small values of u given by the solution of [18], the above implies that:

$$\left|\frac{\beta}{n_1 k} - 1\right| \ll 1 \quad [20]$$

This in turn implies that, for sufficient refractive index change n_2-n_1, w is large and therefore:

$$k_\nu'(w)/k_\nu(w) \simeq 1 \quad [21]$$

Making use of these inequalities in [18] and evaluating the derivatives therein (Abromovitz and Stegun, 1970) results in a simplified eigenvalue equation:

$$\left[\pm v \pm u\frac{J_{v\mp1}(u)}{J_v(u)} + \frac{u^2}{w} \right]\left[\pm v \pm u\frac{J_{v\mp1}(u)}{J_v(u)} + \frac{n_2^2}{n_1^2}\frac{u^2}{w} \right] \simeq v^2 \qquad [22]$$

Expanding the quantity $J_{v\mp1}(u)/uJ_v(u)$ in (negetive) powers of w gives to first order:

$$\frac{J_1(u)}{uJ_0(u)} \simeq \frac{i}{ak\sqrt{n_2^2 - n_1^2}} \qquad [23]$$

$$\frac{J_1(u)}{uJ_0(u)} \simeq \frac{i\,n_2^2}{ak\,n_1^2\sqrt{n_2^2 - n_1^2}} \qquad [24]$$

$$\frac{J_{v\pm1}(u)}{uJ_v(u)} \simeq \frac{\pm i\,(n_2^2 + n_1^2)}{2ak\,n_1^2\sqrt{n_2^2 - n_1^2}} \qquad [25]$$

The equations [23] and [24] correspond respectively to TE and TM modes, whilst [25] corresponds to the EH/HE modes of the dielectric waveguide. Equation [21] implies that the fields are small in the vicinity of the waveguide walls and therefore the particular values of u (and therefore β) that solve the equations above can be found by expanding the Bessel functions about the value of $u^{(o)}$ such that:

$$J_{v\pm1}(u^{(o)}) = 0 \qquad [26]$$

whereupon we find that an approximate solution for each of the roots of [26] is:

$$u^{(1)} \simeq u^{(o)}\left[1 + \frac{i\,b}{ak\sqrt{n_2^2 - n_1^2}} \right] \qquad [27]$$

$$\text{where} \quad b = \begin{cases} 1 & \text{TE modes} \\ n_2^2/n_1^2 & \text{TM modes} \\ (n_2^2 + n_1^2)/2 & \text{EH/HE modes} \end{cases} \qquad [28]$$

We recall from [14] that β is defined in terms of the parameter u:

$$\beta = [n_1^2k^2 - u^2/a^2]^{1/2}$$

$$\simeq n_1k[1 - \tfrac{1}{2}(u^{(1)}/an_1k)^2] \qquad [29]$$

Since [20] requires that β is not much changed from n_1k, the above implies that this analysis is valid only for modes with:

$$u^{(0)} \ll \sqrt{2}an_1k \qquad [30]$$

Of practical import are the losses suffered by the various modes at the waveguide walls. Thus we define the exponential loss coefficient for the electric field intensity:

$$\alpha = -2\,\mathrm{Im}(\beta) \qquad [31]$$

which, from [29], [20], and [27] is found to be:

$$\alpha \simeq 2\left[\frac{u^{(0)}}{ak}\right]^2 \frac{b}{an_1\sqrt{n_2^2-n_1^2}} \qquad [32]$$

2.3 LP modes

Before proceeding to the variational theory for Raman Scattering in a waveguide, it is convenient to make explicit the simplified form for the field components within the framework of the wide bore approximation. Making use of [17] and [20] in equations [8-11] and neglecting terms of the order λ/a, we find for each of the roots of [26] the fields in cylindrical polars are:

$$\underline{\mathrm{TE}} = (\underline{E}, \underline{H}) \sim \exp\left[i\omega t - i\beta z + i\nu\theta\right] J_1\!\left(\frac{ur}{a}\right) J_1^{-1}(u) \left[0, 1, 0, -n_1\sqrt{\frac{\mu_0}{\varepsilon_0}}, 0, 0\right] \quad [33]$$

$$\underline{\mathrm{TM}} = (\underline{E}, \underline{H}) \sim \exp\left[i\omega t - i\beta z + i\nu\theta\right] J_1\!\left(\frac{ur}{a}\right) J_1^{-1}(u) \left[1, 0, 0, 0, n_1\sqrt{\frac{\mu_0}{\varepsilon_0}}, 0\right] \quad [34]$$

$$\underline{\mathrm{EH}}_\nu/\underline{\mathrm{HE}}_\nu = (\underline{E}, \underline{H}) \sim \exp\left[i\omega t - i\beta z + i\nu\theta\right] J_{\nu\pm1}\!\left(\frac{ur}{a}\right) J_{\nu\pm1}^{-1}(u) \left[1, \pm i, 0, \pm i n_1\sqrt{\frac{\mu_0}{\varepsilon_0}}, n_1\sqrt{\frac{\mu_0}{\varepsilon_0}}, 0\right] \quad [35]$$

Having recovered the allowed values for both the real and imaginary parts of β from the eigenvalue equation, it is convenient to pretend that the fields vanish at the waveguide walls and so treat the parameter u in the arguments of the Bessel functions as identically satisfying [26]. Accordingly we drop the normalisation coefficients $J_\nu^{-1}(u)$ (which are effectively absorbed into the field amplitude coefficient).

A further simplification is possible if we note that by taking linear combinations of the form $\frac{1}{2}(\underline{\mathrm{EH}}_{\nu-1} \pm \underline{\mathrm{HE}}_{\nu+1})$ we can construct from [33-35] linearly polarised modes analagous to the TEM_{nm} modes of free-space. Denoting the roots of [26] by $u_{\nu n}$, we define the modes in Cartesian vectors:

$$\underline{\mathrm{LP}}_{\nu n}^{\rightarrow} = \exp\left[i\omega t - i\beta_{\nu n} z + i\nu\theta\right] J_\nu\!\left(\frac{u_{\nu n} r}{a}\right) \left[1, 0, 0, 0, n_1\sqrt{\frac{\varepsilon_0}{\mu_0}}, 0\right] \quad [36]$$

$$\underline{\mathrm{LP}}_{\nu n}^{\uparrow} = \exp\left[i\omega t - i\beta_{\nu n} z + i\nu\theta\right] J_\nu\!\left(\frac{u_{\nu n} r}{a}\right) \left[0, 1, 0, -n_1\sqrt{\frac{\varepsilon_0}{\mu_0}}, 0, 0\right] \quad [37]$$

for the LP modes with x and y polarised electric field components respectively, where β is given by [29]. These are just the linearly polarised modes of the weakly-guiding approximation of fibre-optics (Adams, 1981). Note however that here we do not require $|n_2 - n_1|$ small, but rather the results [36,37] are based instead on the large bore approximation [19].

3. Application to Raman Scattering

3.1 Pump field

Within the framework of section 2, we deduce that any field in the core can be written as a superposition of the LP modes:

$$\{\underline{E}, \underline{H}\} = \mathrm{Re}\left\{ \sum_{\nu n} a_{\nu n}^{\rightarrow} \underline{LP}_{\nu n}^{\rightarrow} + a_{\nu n}^{\uparrow} \underline{LP}_{\nu n}^{\uparrow} \right\} \quad ; \quad r < a \qquad [38]$$

where the field outside the core is supposed to be zero. In the small signal regime, the depletion of the pump beam is negligible and therefore the susceptibilty at the pump frequency is linear. In this case the coefficients $a_{\nu n}$ are constant, and are determined by the field at the entrance to the guide:

$$a_{\nu n}^{\rightarrow} = \frac{\int_0^{2\pi}\int_0^{a} E_x(r,\theta,0,t)\exp(-i\omega t - i\nu\theta)\, J_\nu(U_{\nu n} r/a)\, r\,dr\,d\theta}{\int_0^{2\pi}\int_0^{a} J_\nu^2(U_{\nu n} r/a)\, r\,dr\,d\theta} \qquad [39]$$

and also for the orthogonally polarised fields.

We are now in a position to determine the pump field distribution at the entrance to the guide that in some sense optimises the coupling into the guide modes. In practice, the free-space pump field will be a TEM_{00} beam so that the problem is resolved upon determination of the optimum spot-size and radius of curvature at the guide entrance. The losses indicated by [32] can be shown to be minimised in a quartz guide (n_2=1.5, n_1=1.0) for the HE_{11} mode (Adams, 1980). In our formulation, this means that the LP_{01} mode is the most favoured in terms of the coupling of free-space to waveguide modes. Fortunately, nearly all of the power in a TEM_{00} mode can be coupled into the LP_{01} mode; with the optimally chosen beam parameters, the coupling coefficient can be as high as 0.99 (Abrams, 1975). The optimum TEM_{00} mode has a focus at the guide entrance (z=0) and a spot-size related to the guide bore by:

$$w_p(0)/a = \mu = 0.66 \qquad [40]$$

With these focussing conditions, we will have a pump field in the waveguide which propagates almost entirely as an LP_{01} mode with minimum consequent losses. Using (Abromovitz and Stegun, 1970) $u_{01} = 2.405$, with $n_2 = 1.5$, $n_1 = 1.0$ for a quartz capillary, and recalling:

$$\alpha = \frac{\left(u_{01}^{(0)}\right)^2 \left(n_2^2 + n_1^2\right)}{k^2 a^3 \sqrt{n_2^2 - n_1^2}} \qquad \text{(metres}^{-1}\text{)} \qquad [41]$$

we find the minimum losses for the pump field are:

$$\alpha_p = 0.43\lambda_p^2/a^3 \qquad [42]$$

We will assume henceforth that the free-space pump field is a TEM_{00} mode and the focussing conditions are such that [40] is obeyed whereupon the pump field in the capillary is very nearly an LP_{01} mode with exponential loss given by [42].

3.2 Stokes field

The Stokes field no longer satisfies equation [1] but instead is perturbed in the guide by the presence of the pump induced Raman polarisation. The magnitude of the polarisation term is proportional to the pump intensity which, for the LP_{01} mode, is:

$$I_p = I_{p0} \exp[-\alpha z]\left[J_0\left(\frac{u_{01} r}{a}\right)\right]^2 \qquad [43]$$

Just as in previous chapters, we find the the Raman polarisation induces an effective refractive index at the Stokes frequency that varies both radially and longitudinally. We now use the fact that the projection of the $LP_{\nu n}$ modes onto any one dimension form a complete set of functions within the core of the waveguide. This is true of course (for a fixed polarisation of the LP mode) for either the transverse electric field, or the transverse magnetic field, but not for both. However, it is clear that the Maxwell relation (i.e. equation [6]) fixes the relationship between the transverse magnetic and electric components within each mode, even in the presence of an inhomogeneous electric susceptibility. Therefore the *perturbed* modes of the waveguide must retain the same relationship between the field components, so that provided we remember that the magnetic field components of the Stokes field are given by [6], we can concentrate solely on the electric field. It follows from this discussion that the completeness of the $LP_{\nu n}$ modes can be extended to two dimensions by allowing the coefficients $a_{\nu n}^{\rightarrow}$ and $a_{\nu n}^{\uparrow}$ to become functions of z. Combining $a_{\nu n}^{\rightarrow}$ and $a_{\nu n}^{\uparrow}$ into a single (transverse) vector $\underline{a}_{\nu n}(z) \equiv (a_{\nu n}^{\rightarrow}, a_{\nu n}^{\uparrow})$ we therefore have:

$$\underline{E}_s = R_e \left\{ \sum_{\nu, n} \underline{a}_{\nu n}(z) \exp\left[-i\beta_{\nu n} z + i\nu\theta\right] J_\nu\left(\frac{u_{\nu n} r}{a}\right)\right\} \qquad [44]$$

where $\underline{E}_s$ is a complex amplitude of the Fourier exponential, i.e. in this chapter, the total electric field is:

$$\underline{E} = \mathrm{Re}\ \{\underline{E}_s \exp(i\omega_s t) + \underline{E}_p \exp(i\omega_p t)\}$$

It is easily seen that the form given by [44] is both sufficiently general, and satisfies the appropriate boundary condition, for a Stokes field propagating in a Raman active medium with a pump intensity which is a function of both r and z. Due to the anisotropic nature of the 'susceptibility matrix' (see chapter 1, section 3), we need only consider the Stokes field with a polarisation colinear with that of the pump field. However, note that the orthogonally polarised component will always have a magnitude at least of the order λ_s/a times that of the principle component by virtue of the residual terms neglected in the LP formulation of the waveguide modes.

In order to determine the coefficients $\underline{a}_{\nu n}(z)$, we once again adopt a variational approach based upon the Lagrangian density of chapter 1:

$$\mathcal{L} = \frac{-1}{2\mu_0}\left[\left(\frac{\partial \underline{A}}{\partial x}\right)^2 + \left(\frac{\partial \underline{A}}{\partial y}\right)^2 + \left(\frac{\partial \underline{A}}{\partial z}\right)^2 - \frac{1}{c^2}\left(\frac{\partial \underline{A}}{\partial t}\right)^2 - \left(\underline{\nabla}\,\underline{A}\right)^2 + \frac{\underline{P}}{\varepsilon_0 c^2}\,\frac{\partial \underline{A}}{\partial t}\right] \qquad [45]$$

where $\underline{E} = -\delta\underline{A}/\delta t$ and $\underline{H} = \mu_0\underline{\nabla}X\underline{A}$ [46]

and the polarsiation at the Stokes frequency, is (section 3, chapter 1):

$$\underline{P}_s = \epsilon_0(n_s^2-1)\underline{E}_s + (3/2)\epsilon_0\underline{\underline{\chi}}^{(R)}|\underline{E}_p|^2\underline{E}_s; \quad \underline{P} = \mathrm{Re}\ \{\underline{P}_s\exp(i\omega_s t)\} \qquad [47]$$

where $\underline{\underline{\chi}}^{(R)}$ is a diagonal matrix representing the combined effects of the Raman Scattering susceptibility tensor and the polarisation of the pump field. Note that [44] with $\underline{a}_{\nu n}$ constant finds the minimum of the Lagrangian associated with [45] when the pump intensity is zero.

Writing [45] in cylindrical polars and invoking the zero divergence approximation (chapter 1, section 4) gives the Lagrangian density for the electric field at the Stokes frequency:

$$\mathcal{L} = \frac{-1}{4\mu_0\omega_s^2}\left[\left|\frac{\partial \underline{E}_s}{\partial r}\right|^2 + \frac{1}{r^2}\left|\frac{\partial \underline{E}_s}{\partial \theta}\right|^2 + \left|\frac{\partial \underline{E}_s}{\partial z}\right|^2 - \left|\frac{n_s\omega_s}{c}\underline{E}_s\right|^2 - \frac{3\omega_s^2|\underline{E}_p|^2}{4c^2}\left[\underline{E}_s^*\underline{\underline{\chi}}^{(R)}\underline{E}_s + \underline{E}_s\underline{\underline{\chi}}^{(R)}\underline{E}_s^*\right]\right] \qquad [48]$$

We now proceed noting that terms varying as $\exp(2i\omega t)$ and $\exp(i(\nu-\mu)\theta)$ $(\nu\neq\mu)$ vanish on integration, and assume that we need only retain the component of the Stokes field polarised colinearly with the pump (taken to be in the x direction for definiteness). Thus upon substitution of [43] and [44] into the above, and using [29], we obtain:

$$L = \frac{-\pi}{2\mu_0\omega_s^2} \int dt \int_0^z dz' \Big\{ \sum_{\nu n} \Big[\int_0^a dr\, r\, J_{\nu n}^2\Big(\frac{u_{\nu n} r}{a}\Big)\Big] \cdot \Big[\Big|\frac{\partial a_{\vec{\nu n}}}{\partial z}\Big|^2 + 2i\beta_{\nu n} \mathrm{Re}\Big\{ a^*_{\vec{\nu n}} \frac{\partial a_{\vec{\nu n}}}{\partial z}\Big\}\Big]$$

$$\sum_{\nu nm} a^+_{\vec{\nu n}} \cdot a_{\vec{\nu m}} \Big[\int_0^a dr\, r\, J_{\nu n}\Big(\frac{u_{\nu n} r}{a}\Big) J_{\nu m}\Big(\frac{u_{\nu m} r}{a}\Big) J_0^2\Big(\frac{u_{01} r}{a}\Big)\Big] \cdot \Big[\frac{3\omega^2}{2c^2} |E_{p0}|^2 \chi^{(R)}_{xxxx} e^{-\gamma z' + i(\beta_{\nu n} - \beta_{\nu m}) z'}\Big]\Big\} \qquad [49]$$

We now define the constant C^{ν}_{nm} which is the matrix element of the perturbing potential induced by the pump field:

$$C^{\nu}_{nm} = \frac{\int_0^a dr\, r\, J_{\nu n}\Big(\frac{u_{\nu n} r}{a}\Big) J_{\nu m}\Big(\frac{u_{\nu m} r}{a}\Big) J_0^2\Big(\frac{u_{01} r}{a}\Big)}{\Big[\int_0^a dr\, r\, J_{\nu n}^2\Big(\frac{u_{\nu n} r}{a}\Big) \int_0^a dr\, r\, J_{\nu m}^2\Big(\frac{u_{\nu m} r}{a}\Big)\Big]^{1/2}} \qquad [50]$$

where $$\int_0^a dr\, r\, J_{\nu n}^2\Big(\frac{u_{\nu n} r}{a}\Big) = \frac{1}{2}\Big[\frac{J_{\nu-1}(u_{\nu n})\, a}{u_{\nu n}}\Big]^2 \qquad [51]$$

i.e. $$C_{nm} = 2 u_{\nu n} u_{\nu m} J^{-1}_{\nu-1}(u_{\nu n}) J^{-1}_{\nu-1}(u_{\nu m}) \int_0^1 dx\, x\, J_{\nu n}(u_{\nu n} x) J_{\nu m}(u_{\nu m} x) J_{01}^2(u_{01} x)$$

The matrix C^0_{nm} for n,m $\leqslant$ 10 corresponding to the coupling coefficients for the circularly symmetric modes has been evaluated numerically using an extended (adaptive) Simpson's rule for the integration, and a successive approximation procedure for the Bessel functions (Abromovitz and Stegun, 1970). The results are given in table 3-1. From table 3-1 we find that $\{C^0_{nm}\}$ is a band matrix indicating that coupling takes place only between 'adjacent' modes. Note also that the 'overlap integral' C^0_{11} = 0.566 is close to the value that would be achieved if the pump and Stokes fields were both TEM_{00} Gaussian beams (this is easily found to be 0.5).

The zeroes u_{0m} of the Bessel function $J_0(x)$ have also been found numerically and these are given in table 3-2. The numerical technique employed was the Newton method seeded by an algebraic approximation for each zero (Abromovitz and Stegun, 1970), and once again a successive approximation technique for the Bessel functions.

Upon substitution of [50] into [49] and making use of the paraxial approximation (chapter 1, section 6) we obtain the Euler-Lagrange equations for the coefficient $a_{\vec{\nu n}}(z)$:

$$2i\beta_{\nu n}\frac{da_{\nu n}^{\rightarrow}}{dz} \simeq \frac{3}{2}\frac{\omega_s^2}{c^2}\chi_{xx}^{(R)}|E_{p0}|^2\sum_m a_{\nu m}^{\rightarrow} C_{nm}^{\nu}\frac{J_{\nu-1}(u_{\nu m})}{J_{\nu-1}(u_{\nu n})}\exp\left[i(\beta_{\nu n}-\beta_{\nu m})z-\alpha_p z\right] \quad [52]$$

It is clear from the above that the matrix C_{nm}^{ν} introduces coupling between each of the LP modes at the Stokes frequency. We can employ however a simplifying approximation which consists of ignoring inter-mode but not intra-mode coupling. With reference to the discussion in chapter 2, it was clear from the work of Perry et al (1982) that this approximation was not applicable to the TEM_{nm} free-space modes. But it will be appreciated that this was because the inter-mode phase coherence length could be as great as the effective 'interaction length' between the pump and Stokes fields. In a waveguide however, the interaction length (nominally l) could be much greater than the coherence length (nominally $2\pi/(\beta_{\nu n}-\beta_{\nu m})$, $n\neq m$). Therefore we conclude that *if* the gain for each of the waveguide modes at the Stokes frequency is sufficiently small (i.e. $G_\nu \ll k_s$) *then* we are justified in retaining only the diagonal terms of the scattering matrix $\{C_{nm}^{\nu}\}$ in [52] *provided*:

for all v, m ≠ n: $\quad l \gg 2\pi|\beta_{\nu m} - \beta_{\nu n}|^{-1} \quad (m \neq n)$ [53]

Also noting that:

for all v, m ≠ n: $\quad \mathrm{Min}|u_{\nu m}^2 - u_{\nu n}^2| = |u_{02}^2 - u_{01}^2| = 24.688$ [54]

with reference to [14], we find that [53] and [54] require that the waveguide is long enough to satisfy:

$$a^2/l\lambda_s \ll 0.3 \quad [55]$$

which is easily achieved in the experimental conditions under investigation. Within the framework of this 'long waveguide' approximation we may integrate the now uncoupled equations in [52] to give:

$$a_{\nu n}(z) \simeq a_{\nu n}(0)\exp[\tfrac{1}{2}G_{\nu n}Z_{eff}] \quad [56]$$

where $\quad G_{\nu n} = (3/2)\,\mathrm{Im}\{\chi_{xx}^{(R)}\}k_s|E_{p0}|^2 C_{nn}^{\nu}/n_1$ [57]

is the exponential gain, and

$$Z_{eff} \quad = \quad [1 - \exp(-\alpha_p z)]/\alpha_p \qquad [58]$$

is the effective length of the medium, where α_p is the loss coefficient for the pump beam and we have used [19]. Generally the pump power will be fixed and therefore it is convenient to express the above using [39] as:

$$G_{\nu n} \quad = \quad \check{C}_{nn}\tilde{P}_p/(\mu^2 a^2 k_s) \qquad [59]$$

where $$\tilde{P}_p \quad = \quad (3/2)\,\mathrm{Im}\{\chi^{(R)}_{xx}\}k_s^2|E_{po}|^2 w_{po}^2/n_1 \qquad [60]$$

is the normalised pump power introduced in chapter 2.

Note from table 3-1 that the growth $G_{\nu n}$ is a maximum for the LP_{01} mode whence $\check{C}_{nn} = 0.566$. Thus the pump mode that optimally couples into the guide (TEM_{00}/LP_{01}) also maximises the growth of a single (circularly symmetric) Stokes mode within the guide (i.e. the LP_{01} mode).

From the analysis of this section it is clear that we are able to treat the Stokes field as a series of independent modes growing at a rate given by equations [56] and [57]. The net gain experienced by each mode however, is a combination of the Raman gain and the losses suffered on reflection at the waveguide walls. Recalling equation [44] then, we can write the full Stokes field as:

$$\underline{E}_s \quad \simeq \quad \Sigma_{\nu n}\, a_{\vec{\nu n}}(0) J_{\nu n}(u_{\nu n} r/a)\exp[\; i\nu\theta + i\omega_s t - ik_s z + \tfrac{1}{2}\gamma_{\nu n} \;]\underline{e}_x \quad [61]$$

where $$\gamma_{\nu n} \quad = \quad [\; G_{\nu n} Z_{eff} - \alpha_s z \;] \qquad [62]$$

is the net Stokes exponential gain, and $a_{\vec{\nu n}}(0)$ is the (supposed) injected amplitude, of the $LP_{\nu n}$ mode polarised colinear with the pump field, and α_s is the exponential loss at the Stokes frequency. This result has been anticipated by Berry and Hanna (1983) and used to obtain predictions in good agreement with experimental results.

4. Optimum guide characterisitics

Before we can assess the efficacy of the waveguide, say in comparison to the focussed pump beam method of chapter 2, it is necessary to consider the effects on the Stokes gain of varying the parameters at the experimentalist's disposal. Once again, we are mostly concerned with the choice of parameters that minimises the pump power necessary to attain some fixed threshold. In this section then, we will show how the variation of these parameters affects the net Stokes gain, so that an optimal choice can be made consistent with constraints on the design of a particular Raman laser.

The gross behaviour of the net Stokes gain may be determined from equations [58,59] and [62]. The main feature is the competition between the effects of guidance and enhancement through $G_{\nu n}$ - which increases for small bore radius (equation [59]) - and the losses suffered at the waveguide walls through α_s and Z_{eff} - which for large radii are less important, but which increase rapidly as $a \to 0$. Clearly therefore, for all other parameters constant, the net gain has a distinct maximum for the optimal choice of bore radius.

The situation can be further clarified by reference to a graphical representation of [62]. Firstly we define a set of normalised co-ordinates $\{x_1, x_2, x_3\}$ which serve as a vehicle for expressing the degrees of freedom available in the design of a waveguide laser.

Let $\quad x_3 \;=\; k^2\gamma_{\nu n} \;; \qquad k = \; k_s/k_p$ [63]

be the net exponential Stokes gain (scaled by ^{2}),

$$x_2{}^3 \;=\; \alpha_p l \qquad [64]$$

be the (exponential) pump losses, and

$$x_1 \;=\; k^2 G_{\nu n} x_2/\alpha_p \qquad [65]$$

is the new normalised pump power. Then [62] can be written:

$$x_3 = x_1[\ 1 - \exp(-x_2^3)]/x_2 - x_2^3 \qquad [66]$$

The choice of definitions is motivated by the need to decouple the dependencies of the $\{x_i\}$ on the intrinsic design variables which are here taken to be guide bore and length. Thus only x_2 depends on a, and whilst both x_1 and x_2 depend on l, the mutual coupling is linear, so that a straight line through the origin in the (x_1,x_2) plane represents the locus of x_1,x_2 for varying l and fixed a.

In terms of these parameters, for the LP_{01} mode in a fused silica guide, with $n_1 = 1$ in the core, the $\{x_i\}$ are:

$$x_1 = 0.36\breve{P}_p(\ \lambda_p^2 l/\lambda_s^3\ )^{1/3} \qquad [67]$$

$$x_2 = 0.75(\ \lambda_p^2 l/a^3\ )^{1/3} \qquad [68]$$

$$x_3 = \gamma_{01}(\ \lambda_p^2/\lambda_s^2\ ) \qquad [69]$$

With these definitions, the projection in the (x_1,x_2) plane of various values of x_3 given by [66] is given in figure 2. With reference to figure 2, we can make the following observations:

1) The net Stokes gain increases monotonically with pump power for fixed l, a.

2) The net Stokes gain has a single maximum at some bore radius for fixed $\breve{P}_p$, l.

3) The maximum net Stokes gain (for optimal a) is a monotonicly increasing function of l for fixed $\breve{P}_p$.

4) The pump power required to attain some net Stokes gain is a monotonically increasing function of that Stokes gain for fixed l, a.

5) The pump power required to attain some net Stokes gain has a single minimum at some bore radius for fixed $\breve{P}_p$, l.

6) The minimum pump power required to attain some net Stokes gain (for optimal a) is a monotonically decreasing function of l.

More specifically, we can outline a set of procedures using figure 2, for various optimisation schemes as follows:

a) Find the optimal bore radius and length that maximise the Stokes gain for fixed pump power:

From the third of the points above, it is clear that the maximal Stokes gain increases without limit with increasing l. This suggests that a guide of the maximum tolerable length l_{max} is chosen. In practice it will not be just the physical dimensions of the guide that detemine l_{max}. We note from figure 2 that the optimal choice of bore becomes progressively more critical with increasing l. Also, the losses suffered from (as yet unaccounted for) imperfections in the guide, including bends, will undoubtedly take their toll as l increases. Given l_{max}, we can determine $x_1(max)$ from [67]. Then a line $x_1 = x_1(max)$ in figure 2 determines $x_3(max)$ and therefore the optimal bore via x_2.

b) Find the optimal length of the guide that maximises the net Stokes gain for fixed pump power and bore radius.

In this case we can draw a line through the origin of the (x_1,x_2) plane at an angle $\theta = \tan^{-1}(x_2/x_1)$ which according to equations [67] and [68] is $\tan^{-1}(2\lambda_s/(a\breve{P}_p))$. The point on the line that maximises x_3 detemines the co-ordinates (x_2,x_1) and therefore l. It is easy to show from [66] and definitions [67-69] that the optimal length is:

$$l_{opt} = (lx_2^{-3})\ln(x_1/x_2) \qquad [70]$$

i.e. $$l_{opt} \simeq 2.4(a^3/\lambda_p^2)\ln(a\breve{P}_p/(2\lambda_s)) \qquad [71]$$

It is clear from figure 2 that in most cases, unless the pump losses are very high, the net Stokes gain continues to increase with l beyond $x_3 = 45$ when the bore radius is fixed. Although [71] suggests that there is always a distinct optimal length, once again design constraints may dictate a maximum tolerable length before this value is reached.

c) Find the minimum pump power necessary to attain a fixed threshold for arbitrary guide bore and length.

Here we have x_3 given, and x_1, x_2 to be determined. With reference to the sixth point above, it is clear that we must again choose l to be the maximum tolerable length. With reference to figure 2, the minimal pump power is then determined from the point of intersection of the line x_1 = constant which is a tangent to the curve x_3 = constant (given).

d) Find the minimum pump power necessary to attain a fixed threshold given a guide of fixed bore radius of arbitrary length.

From point b) above, we note that the angle θ of a line through the origin in the (x_1,x_2) plane is a monotonically decreasing function of $\hat{P}_p$. Therefore we seek the maximum angle of such a line that passes through the curve x_3 = constant (given). This line is just the tangent to the curve x_3 = constant, the point of intersection determining the optimal co-ordinates (x_1,x_2) and therefore the minimal pump power and optimal bore radius. It may be verified with reference to figure 2, that this optimisation procedure is consistent with the approximation:

$$\gamma_{01} \approx G_{01}/\alpha_p \qquad [72]$$

which in normalised co-ordinates is:

$$x_3 \approx x_1/x_2 \qquad [73]$$

Thus within this optimisation scheme, the net Stokes gain for the LP_{01} mode is approximately:

$$\gamma_{01} \approx (\breve{P}_p/2\breve{k}).(a/\lambda_p) \qquad [74]$$

We conclude this chapter with an example comparing free-space and guided Raman Scattering. At least for the scheme d) above, and recalling the results of chapter 2 for Stokes gain using a focussed pump beam, we note that [74] predicts a reduction by a factor of order $a/(\lambda_p\pi)$ in the pump power necessary to attain threshold in a waveguide. However, this is only a very approximate comparison, so that we now proceed with a hypothetical example as follows.

Suppose that we require the minimum pump power required to reach a threshold Stokes intensity given by a gain factor of exp(40) for Raman Scattering in H_2 gas with $\lambda_p = 1.06\mu m$ whereupon the first Stokes has a wavelength $\lambda_s = 1.91\mu m$. Let us also suppose that practical design considerations dictate that $l_{max} = 1m$. In the tight-focussing high pump-power regime of free-space generation, we have from chapter 2:

$$\gamma = \pi(\breve{P}_p - 2\sqrt{\breve{P}_p})/(2k) \qquad [75]$$

which with $\gamma = 40$ and $k = 0.55$ gives $\breve{P}_p \approx 24$ for threshold. (This value confirms our supposition that we are working in the domain of high pump power).

In the waveguide, for the LP_{01} mode we have $x_3 = k^2\gamma_{01} \approx 12$. With reference to figure 2, and executing procedure c) above, we find the intersection co-ordinates to be $(x_1,x_2) \approx (17,1.0)$. The value for x_2 gives the optimal bore radius from [68] to be $a \approx 80\mu m$. The value for x_1 gives the minimal pump power from [67] to be $\hat{P}_p \approx 1$ for threshold.

Thus we conclude that, even in consideration of the design constraints on the choice of guide length, Raman Scattering in a waveguide confers a significant advantage in terms of reducing pump threshold power compared with Raman Scattering in free-space using a focussed pump beam.

Table 3-1

Matrix elements (C_{nm}) for the circularly symmetric modes

n\m	1	2	3	4	5	6	7	8	9	10
1	0.566	0.271	0.013	-0.001						
2	0.271	0.485	0.256	0.013	-0.002					
3	0.013	0.256	0.478	0.253	0.013	-0.002				
4	-0.001	0.013	0.253	0.476	0.253	0.013	-0.002			
5		-0.002	0.013	0.253	0.475	0.252	0.013	-0.002		
6			-0.002	0.013	0.252	0.474	0.252	0.013	-0.002	
7				-0.002	0.013	0.252	0.474	0.252	0.013	-0.002
8					-0.002	0.013	0.252	0.474	0.252	0.013
9						-0.002	0.013	0.252	0.474	0.252
10							-0.002	0.013	0.252	0.474

Table 3-2

Zeroes of $J_0(x)$ for $x>0$

n	u_{0n}
1	2.405
2	5.520
3	8.654
4	11.792
5	14.931
6	18.071
7	21.212
8	24.352
9	27.493
10	30.635

FIGURE 1

Co-ordinate system and parameters associated with the waveguide

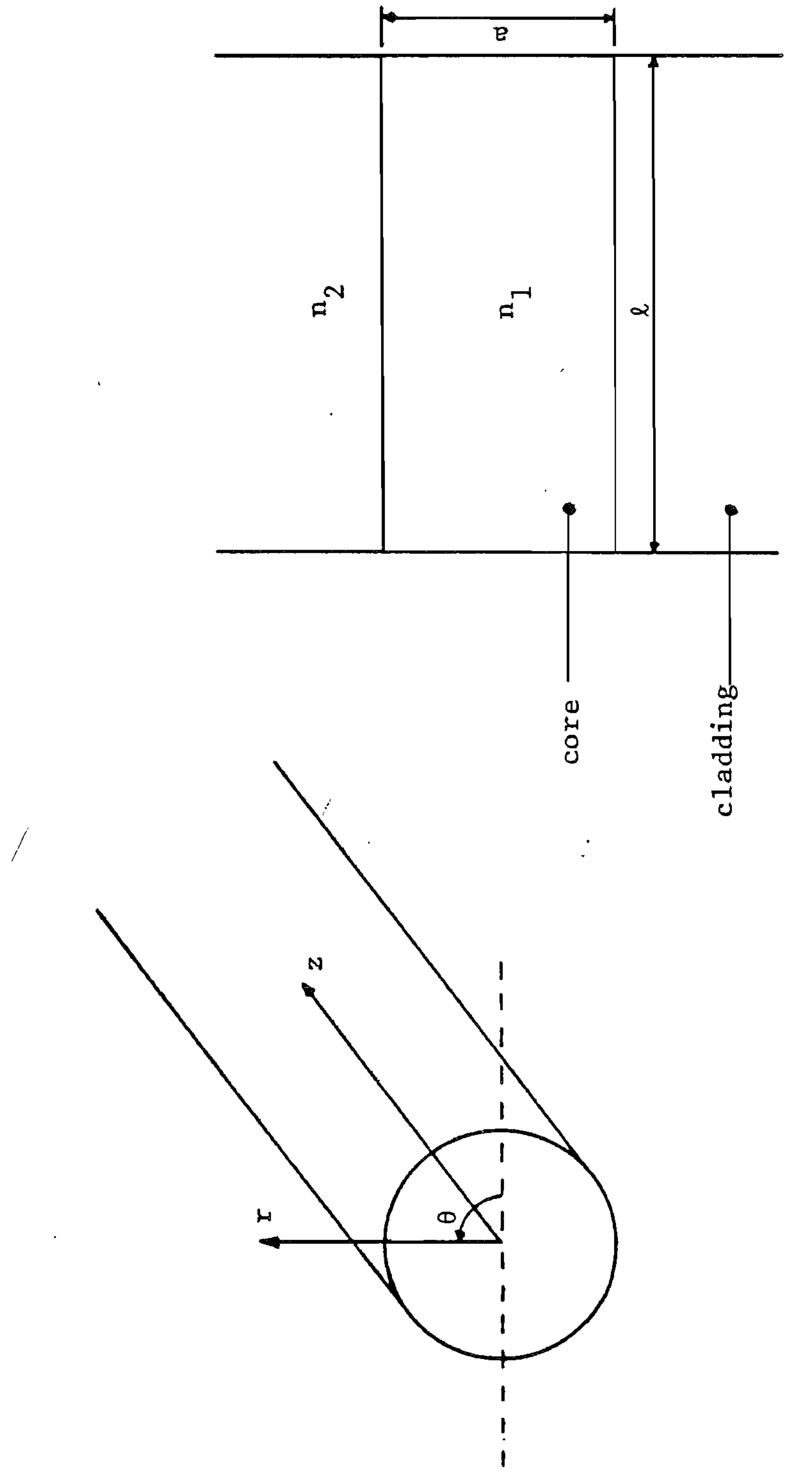
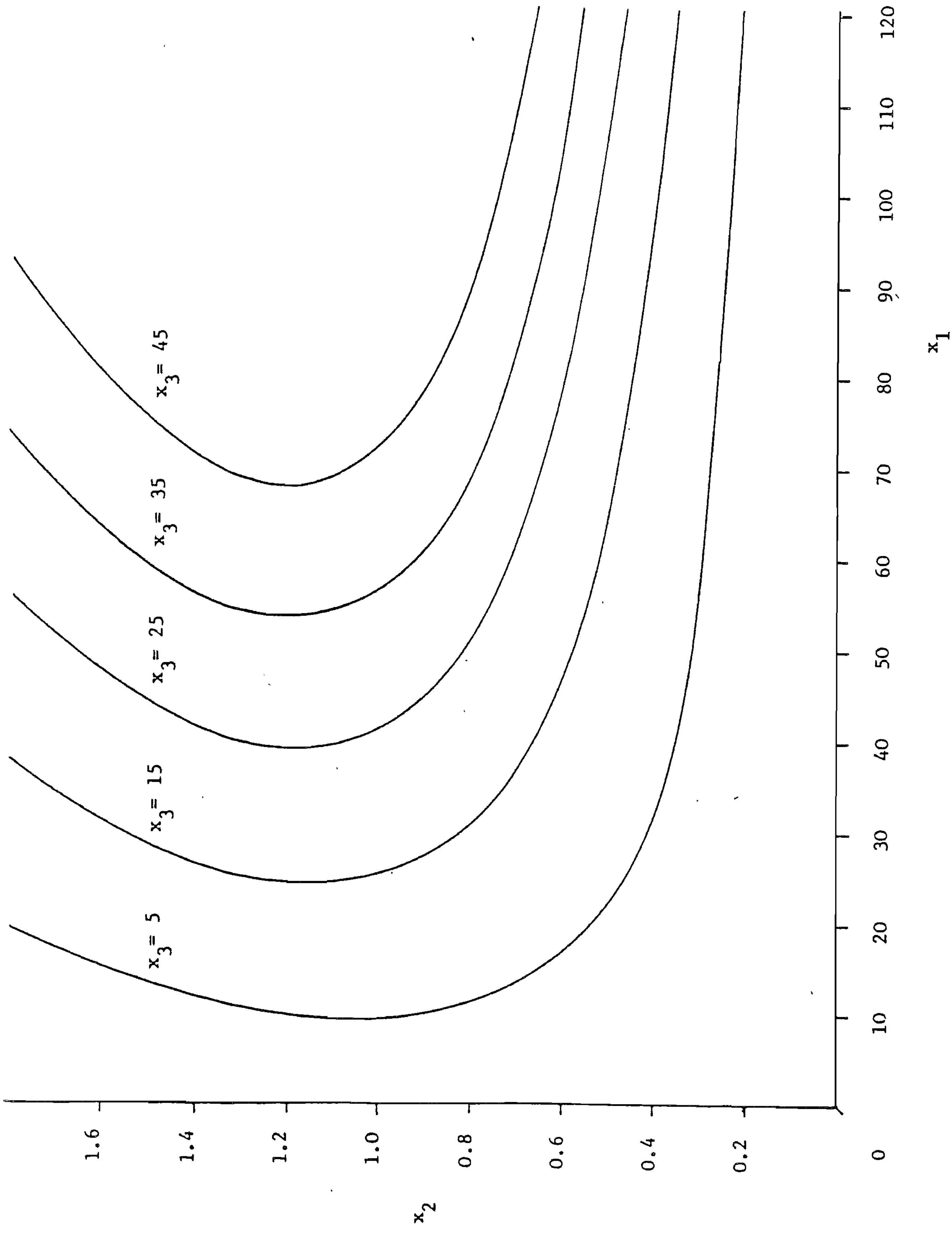


FIGURE 2

Curves of constant normalised gain (x_3) with normalised pump power (x_1) and pump losses (x_2) for a Stokes beam in a Capillary Waveguide.



CHAPTER 4

RAMAN SCATTERING WITH A TIME VARYING PUMP BEAM

1. Introduction

The Stimulated Raman Scattering steady-state plane wave gain coefficient G_R, discussed in chapter 1, section 7, depends on the pump field only through the intensity I_p. If the pump laser is pulsed (rather than c.w.), then the maximum energy in each pulse will be determined by the output of the flash-lamp and the efficiency of the laser. Thus, it would seem possible to control - and therefore increase - the value of G_R through control of the pump laser pulse width. If the gain coefficient is to be maximised, then we will need to know the extent to which the steady-state result - and therefore this line of reasoning - can be trusted. We will also need a more general theory capable of describing the behaviour of a Stokes field when the Stimulated Raman Scattering process is no longer steady-state. Much work has already been done in this field, and so leaving aside the technical aspects of short pulse generation, we will concentrate on some of the more neglected aspects of the problem.

It is possible to identify two extreme regions in the time dependent behaviour of the Stokes field which are analagous to the plane-wave and tightly focussed domains of free-space Raman Scattering (chapter 2). If the steady-state regime is likened to the plane-wave case of that chapter, then the tightly focussed pump beam (i.e. spatial confinement) may be likened to a transient limit for short pump pulses (temporal confinement). Of very general interest, we investigate in this chapter the advantages offered by a system that is neither steady-state, nor plane-wave. In particular, we will use some of the ideas introduced in chapters 2 and 3, and some of the familiar results from the field of time dependent Raman Scattering, to describe the behaviour of the Stokes field in a medium excited by a short, focussed or guided, pump pulse.

2. Maxwell-Bloch equations

In this work, we make use of the semi-classical model of Stimulated Raman Scattering which is discussed for instance in Yariv (1975). The full quantum mechanical treatment has been given in Penzkoffer et al (1979). Perhaps more usefully, Raymer and Mostowski (1981), have restricted the quantisation of the electromagnetic fields to that of the pump laser, while considering in some detail the spontaneous initiation of the Stokes field. In chapter 1, section 3, it was assumed that the classical third order susceptibility was sufficient to describe the polarisation of the medium at the Stokes frequency:

$$\underline{P}_s = \epsilon_o(n_s^2-1)\underline{E}_s + (3/2)\epsilon_o\chi^{(3)}(-\omega_s;\omega_p,-\omega_p,\omega_s)\underline{E}_p\underline{E}_p^*\underline{E}_s \qquad [1]$$

In this chapter, we assume that the Stokes field is polarised colinear with the pump (taken for definiteness in the x direction) and therefore retain the scalar formulation of the field equations. Equation [1] however is valid only in the steady-state limit; for a more general description of the time dependent behavior of the Stokes field, we must return to the molecular oscillator, and its interaction with a time varying pump field.

In our semi-classical description, we let Q be the oscillator coordinate identifiable with the expectation value of the quantum mechanical dipole operator. Raman Scattering takes place when the polarisability α, is a function of the dipole length; i.e. for each of the N oscillators:

$$\alpha = \alpha_o + Q(\delta\alpha/\delta Q)|_{Q=0} \qquad [2]$$

where the susceptibility $\chi = N\alpha$ and N is the number of oscillators within the region under consideration. Thus the energy density of the dipoles in an electromagnetic field is

$$\tfrac{1}{2}(1+\chi)\epsilon_o E^2 = \tfrac{1}{2}\epsilon_o(1+N(\alpha_o + Q(\delta\alpha/\delta Q)|_{Q=0}))E^2 \qquad [3]$$

(Here we are working with scalar fields; the extension Q is taken to be measured along the direction of the field vector $\underline{E}$.)

The force exerted by the field on each oscillator is therefore:

$$F = \tfrac{1}{2}\epsilon_o(\delta\alpha/\delta Q)E^2 \qquad [4]$$

Hence the coordinate Q is modelled as satisfying an harmonic oscillator equation with a forcing term proportional to [4]:

$$\frac{d^2Q}{dt^2} + 2\Gamma_A\frac{dQ}{dt} + \omega_v^2 Q = \frac{\varepsilon_o}{2m}\frac{\partial\alpha}{\partial Q}E^2 \qquad [5]$$

where ω_v is the resonant frequency of the oscillator, m its effective mass, and Γ_A is the collision induced damping. The reader is reminded that this approach is justified only when the fractional change in the population of the quantum mechanical ground state is small, whence the Heisenburg equations of motion for the dipole operator are linear in the forcing term given by [4]. Thus equation [5] becomes invalid when the scattering starts to appreciably deplete the active medium. Note also that the spatial derivatives that would be present in a wave equation involving Q have been ignored in this treatment. This neglect is justified by the relatively low velocity of the (optically induced) phonons in the Raman active medium (which in our case is a gas), i.e. each oscillator can be considered as uncoupled and independent from its neighbours.

If we now suppose that the field is composed of mainly two components E_p and E_s at frequencies ω_p and $\omega_s = \omega_p - \omega_v$ respectively, then the oscillator is driven most effectively by the cross terms at the resonant frequency. As in previous cases, we extract the major components of the time and space variation whilst leaving a slowly varying amplitude to absorb any deviation from a purely sinusoidal behaviour:

$$Q = \mathrm{Re}\left\{ q\, e^{i\omega_v t} \right\}$$

$$E = \mathrm{Re}\left\{ \epsilon_s\, e^{i\omega_s t - ik_s z} + \epsilon_p\, e^{i\omega_p t - ik_p z} \right\}$$

where $k_s = \omega_s n_s/c$, $k_p = \omega_p n_p/c$, and $\omega_v \gg \Gamma_A$. [6]

The equation of motion for the slowly varying amplitude q is now:

$$dq/dt + \Gamma_A q \simeq -ik_1{}^*\epsilon_p\epsilon_s{}^* \qquad [7]$$

where $k_1{}^* = (\delta\alpha/\delta Q)|_{q=0}.\epsilon_o/(8m\omega_v)$ [8]

and corresponds to the (quantum mechanically derivable) coupling constant (Penzkoffer et al 1979):

$$K_1 = \sum_m \frac{\mu_{im}\mu_{mg}}{\hbar^2}\left[\frac{1}{\omega_{mg}-\omega_p} + \frac{1}{\omega_{mg}+\omega_s}\right] \qquad [9]$$

Note that the steady-state limit is just that of $dq/dt \to 0$ whence:

$$q = -i(k_1{}^*/\Gamma_A)\epsilon_p\epsilon_s{}^* \qquad [10]$$

is constant in time.

ϵ_p and ϵ_s may contain spectral components with a combined bandwidth much greater than Γ_A whilst retaining their status as slowly varying envelopes for the optical frequencies ω_p and ω_s respectively. Thus [7] is a useful starting point for our discussion of the temporal dependence of the stimulated process. We now follow Raymer and Mostowski (1981) and introduce a Langevin function $F(\underline{r},t)$ that is responsible for the collision induced fluctuations:

$$dq/dt + \Gamma_A q = -ik_1{}^*\epsilon_p\epsilon_s{}^* + F \qquad [11]$$

Unlike Raymer et al however, our function will be used to describe initiation of the scattering process in the volume defined by the active medium. Thus we have:

$$\langle F(\underline{r}',t')F^*(\underline{r},t)\rangle = 2(\Gamma_A/\rho_v)\delta(\underline{r} - \underline{r}')\delta(t - t')$$

$$\langle q(\underline{r}',0)q^*(\underline{r},0)\rangle = (1/\rho_v)\delta(\underline{r} - \underline{r}') \qquad [12]$$

and ρ_v is the molecular volume density.

The Langevin force is introduced in anticipation of the need for a source capable of initiating the scattering process when there is no electromagnetic field injected at the Stokes frequency. Once within the stimulated regime, the quantative bevaiour of the Stokes field is almost indifferent to the type of process, spontaneous or otherwise, assumed responsible for the initiation. The precise definition of F should therefore not be taken too seriously; (there are for example difficulties in assuming that the fields can be written as a slowly varying envelope times a Fourier component whilst F obeys [12]!).

It now remains only to describe how each oscillator contributes to the total polarisation of the medium. It can be seen that the non-linear polarisation of the medium depends on q through [2] and [6]. At the Stokes frequency, the polarisation will involve products of q and ϵ_p:

$$P_s^{(NL)} \;\; \alpha \;\; q^*\epsilon_p \exp(i(\omega_s t - k_s z)) \qquad [13]$$

Again we follow the notation of previously published work and define the modified coupling constant:

$$k_2 \;=\; \tfrac{1}{2}\rho_v h\omega_s k_1{}^*/(\epsilon_o c) \qquad [14]$$

The Maxwell equation for the slowly varying amplitude ϵ_s under the paraxial approximation can be written, using [13] and [14]:

$$\left\{ \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2iK_s\left[\frac{\partial}{\partial z} + \frac{n_s}{c}\frac{\partial}{\partial t}\right]\right\}\epsilon_s \;=\; -2K_s K_2 q^* \epsilon_p \qquad [15]$$

Equations [15] and [11] are the coupled Maxwell-Bloch equations for the atom - field interaction. They form the basis for the subsequent discussion in this chapter on the temporal characteristics of Raman Scattering outside the steady-state regime. It is appropriate at this point to note that there is a reciprocal polarisation at the frequency ω_p, which involves products of q and ϵ_s. As in chapter 2, this component is ignored thereby confining our analysis to the small-signal regime.

3. Time dependent behaviour of plane wave Raman Scattering

There has been no published work known to the author which attempts to treat simultaneously both the time dependent, behaviour, and the diffraction/gain-focussing behaviour of the Stokes field. Instead, in modelling the time dependent behaviour, previous authors assume that both the pump and Stokes fields are plane waves, and therefore that the transverse derivatives in [15] can be neglected:

$$\left[\frac{\partial}{\partial z} + \frac{n_s}{c} \frac{\partial}{\partial t} \right] \epsilon_s = -i k_2 q^{+} \epsilon_p \qquad [16]$$

where Raymer and Mostowski have $F(\mathbf{r},t) \rightarrow F(z,t)$, $q(\mathbf{r},t) \rightarrow q(z,t)$, and ρ_1 is now the molecular density per unit length.

Even with this simplified model, there exists a wide range of sub-domains each concerned with a very particular aspect of the time dependent Raman Scattering problem. In this chapter we make an additional assumption that will enable us to concentrate on just a few of the possible instantiations of [11] and [15]. This approximation is that the dispersion of the medium is negligible for the fields at the pump and Stokes frequencies, the validity of which depends on the pump pulse width and the the length of the active medium. Consider for example a pump pulse of width τ_p in an active medium with refractive index at frequencies ω_p, ω_s of n_p, n_s respectively. Then a medium of length greater than:

$$l_{coh} = c\tau_p |n_p - n_s|^{-1} \qquad [17]$$

would not support the use of our dispersionless model; a co-propagating Stokes pulse would not experience the full gain over the full length of the medium. If the active medium is a gas, the material dispersion can be reduced by introduction of an additional gas with material dispersion of opposite sign. If this is not feasible, then a more comprehensive model must be considered; see for instance the discussion by Carman et al (1970), and Akhmanov et al (1971 and 1974).

Under the approximation $n_p \simeq n_s = n$, the Maxwell-Bloch equations for fields in a dispersionless medium simplify through the transformation:

$$\tau = t - nz/c \tag{18}$$

whence [11] and [16] become:

$$\delta\epsilon_s(z,\tau)/\delta z = -ik_2 q^*(z,\tau)\epsilon_p(\tau) \tag{19}$$

$$\delta q(z,\tau)/\delta\tau + \Gamma_A q(z,\tau) = -ik_1{}^*\epsilon_p(\tau)\epsilon_s{}^*(z,\tau) + F(z,\tau) \tag{20}$$

If the pump field envelope is assumed to suffer no variation as it propagates through the medium, ϵ_p is a function of τ only. Upon the substitution:

$$U(z,\tau) = \epsilon_p^{-1}(\tau)\,\epsilon_s(z,\tau)\,e^{\Gamma_A\tau} \tag{21}$$

we obtain from the above:

$$\frac{\partial U}{\partial z} = -iK_2 q^* e^{\Gamma_A\tau} \tag{22}$$

$$\frac{\partial}{\partial\tau}\left[q^* e^{\Gamma_A\tau}\right] = iK_1|\epsilon_p|^2 U + F^* e^{\Gamma_A\tau} \tag{23}$$

These may be combined to give:

$$\frac{\partial^2 U}{\partial z\partial\tau} - K_1K_2|\epsilon_p|^2 U = -iK_2 e^{\Gamma_A\tau}F^* \tag{24}$$

We now assume that the initial fields $q(z,0)$ and $\epsilon_s(0,\tau)$ are given, whence [24] can be solved (Raymer and Mostowski, 1981) to give:

$$\begin{aligned}\epsilon_s(z,\tau) - \epsilon_s(0,\tau) = {}& -iK_2\epsilon_p(\tau)\,e^{-\Gamma_A\tau}\int_0^z dz'\,q^*(z',0)\,I_0\left[4K_1K_2(z-z')\,p(\tau)\right]^{1/2}\\ &+\sqrt{K_1K_2z}\;\epsilon_p(\tau)\int^{\tau} d\tau'\left[p(\tau)-p(\tau')\right]^{-1/2}e^{-\Gamma_A(\tau-\tau')}\epsilon_p^*(\tau')\,\epsilon_s(0,\tau')\,I_1\left[\left[4K_1K_2z\left(p(\tau)-p(\tau')\right)\right]^{1/2}\right]\\ &-iK_2\,\epsilon_p(\tau)\int_0^{\tau}d\tau'\int_0^z dz'\,e^{-\Gamma_A(\tau-\tau')}F^*(z',\tau')\,I_0\left[4K_1K_2(z-z')\left(p(\tau)-p(\tau')\right)\right]^{1/2}\end{aligned} \tag{25}$$

where the $I_n(x)$ are modified Bessel functions, and:

$$p(\tau) = \int_0^{\tau} |\epsilon_p(\tau'')|^2 d\tau'' \qquad [26]$$

is proportional to the energy in the pump pulse upto time τ.

If the Raman Scattering process is deemed to be initiated solely by the fluctuations in $q(z,0)$ and $F(z,\tau)$, then:

$$\langle \epsilon_s(0,\tau')\epsilon_s^*(0,\tau')\rangle = 0 \qquad [27]$$

Using the above, with the auto correlation functions [12], the product

$$I_s(z,\tau) = \tfrac{1}{2}\epsilon_0 c \langle \epsilon_s^*(z,\tau)\epsilon_s(z,\tau)\rangle$$

may be evaluated, and the integrations over z may thus be performed (see Raymer et al, 1981) to give the Stokes intensity (in SI units $Jm^{-2}s^{-1}$):

$$I_s(z,\tau) = \tfrac{1}{2}\varepsilon_0 c\, |\kappa_2|^2/\rho_\ell\, |\epsilon_p(\tau)|^2 z \ .$$

$$\Big\{ e^{-2\Gamma_A \tau} \Big[I_0^2\big[(4\kappa_1\kappa_2 z\, p(\tau))^{1/2}\big] - I_1^2\big[(4\kappa_1\kappa_2 z\, p(\tau))^{1/2}\big]\Big] \qquad [28]$$

$$+ 2\Gamma_A \int_0^{\tau} d\tau'\, e^{-2\Gamma_A(\tau-\tau')}\Big[I_0^2\big[(4\kappa_1\kappa_2 z(p(\tau)-p(\tau')))^{1/2}\big] - I_1^2\big[(4\kappa_1\kappa_2 z(p(\tau)-p(\tau')))^{1/2}\big]\Big]\Big\}$$

4. Steady-state and transient limits

The result [28] is a general description of the time dependent behaviour of the Stokes intensity under the small-signal, plane-wave, zero dispersion conditions assumed to hold in section 3. The *steady-state* plane-wave limit (chapter 1, section 7) can be recovered from [28] by taking the limit as $\Gamma_A \tau \to \infty$. This is justified, given that $\epsilon_p(\tau)$ is constant, by the lack of temporal variations of any consequence in the integrand, whence [28] reduces to:

$$I_s(z,\infty) = \frac{1}{2}\epsilon_0 c|k_2|^2/\rho_L|\epsilon_p|^2 z \int_0^\infty dx\, e^{-x}\left\{ I_0^2\left[\left(\frac{2k_1k_2|\epsilon_p|^2 x}{\Gamma_A}\right)^{1/2}\right] - I_1^2\left[\left(\frac{2k_1k_2|\epsilon_p|^2 x}{\Gamma_A}\right)^{1/2}\right]\right\} \quad [29]$$

which can be integrated to give (Gradshteyn and Ryzik):

$$I_s(z,\infty) = \frac{1}{2}\epsilon_0 c|K_2|^2/\rho_L|\epsilon_p|^2 z\left[I_0\left(\frac{K_1K_2|\epsilon_p|^2 z}{\Gamma_A}\right) - I_1\left(\frac{K_1K_2|\epsilon_p|^2 z}{\Gamma_A}\right)\right]\exp\left[\frac{K_1K_2|\epsilon_p|^2 z}{\Gamma_A}\right] \quad [30]$$

The familiar steady-state result can at last be recovered if the gain is sufficiently high whence the modified Bessel functions can be approximated by taking the first term in the expansion (Abromovitz and Stegun):

$$I_n(x) = (2\pi x)^{-1/2}e^{x}(1 - (4n^2-1)/(8x) + \dots) \text{ for } x \gg 1$$

whence $$I_s(z,\infty) \simeq \frac{\hbar\omega_s\Gamma_A\rho_v}{8\rho_L\sqrt{G_R z\pi}}\exp\left[G_R z\right] \quad [31]$$

where $$G_R = 2k_1k_2|\epsilon_p|^2/\Gamma_A \quad [32]$$

can be identified as the Raman gain coefficient - now expressed in terms of quantities which are (at least in principle) calculable. To cast this into the familiar result of steady-state theory of chapter 1, we let:

$$I_{so}^{(ss)} = \frac{\hbar\omega_s\Gamma_A\rho_v}{8\rho_L\sqrt{G_R z\pi}}$$

which is a weak function of z, and for our purposes can be regarded as a constant, whence:

$$I_s \simeq I_{so}^{(ss)}\exp[G_R z] \quad [33]$$

The validity of this result rests on the (time-wise) dominant behaviour of the exponential factor in the integrand in [28]. For a more detailed assessment of the situation, the reader is referred to equation [20]. This suggests that, for the steady-state result to be accurate, the bandwidth of the product $\epsilon_p(\tau)\epsilon_s^*(z,\tau)$ must be much less than the linewidth Γ_A - a condition which corresponds physically to (adiabatic) following by the molecular oscillator of the driving signal. From the exponential dependency indicated by [31], and the form of the driving term in [20], we can argue that in the high gain limit, the pump field must have a bandwidth Γ_p that satisfies:

$$\Gamma_A \quad > \quad \tfrac{1}{2}G_R z\Gamma_p \qquad [34]$$

Moreover, for the transient component of the Stokes intensity to be negligible, we require that the time elapsed is sufficient such that the first time in [28] is almost zero. It is easy to show that, for a 'top hat' pump pulse of duration τ_p, the first term has a sharp peak at a time

$$\tau \quad \simeq \quad \tfrac{1}{2}G_R z/\Gamma_A \qquad [35]$$

and thereafter rapidly decays to zero. The second term however, continues to increase monotonically after this time. Therefore a time τ_p such that

$$\tau_p \quad > \quad \tfrac{1}{2}G_R z/\Gamma_A \qquad [36]$$

is a *sufficient* minimum pulse length that must be endured before the system settles to the steady-state response; (note that this is just [34] with Γ_p replaced by τ_p^{-1}).

Generally, the transient limit of the result for plane wave Raman Scattering is deemed to occur when the elapsed time of the 'top hat' pump pulse envelope satisfies:

$$\Gamma_A\tau_p \quad \ll \quad 1 \qquad [37]$$

whence the second term in [28] is negligible with respect to the first. If now in addition to [37], the gain is sufficiently great then the Bessel functions can be expanded as for [31], and the high gain limit for transient Raman Scattering recovered:

$$I_s \simeq I_{so}^{(tr)} \exp[2(2G_R z \Gamma_A \tau_p)^{1/2}] \qquad [38]$$

where $$I_{so}^{(tr)} = \frac{\hbar \omega_s \rho_r}{32\pi \rho_L \tau_p}$$

and valid when: $G_R z \Gamma_A \tau_p \gg 1$ [39]

Clearly, for a sufficiently small pump pulse width τ_p, the Stokes field never leaves the transient domain described by [37] and [38].

From the steady-state and transient domain results we can draw the following conclusions. The exponential gain coefficient in the steady-state limit depends only on the pump *intensity*. In the transient limit, the exponential gain coefficient depends only on the *energy* in the pump pulse. Since the gain coefficient is a smooth monotonic function of the pump pulse width (see for instance the results of Raymer et al, 1979), we may conclude that given a fixed energy pump pulse of variable duration, the Raman threshold is most easily achieved using the shortest possible pulse. However, once within the transient domain, there is no advantage in further shortening the pump pulse width. Thus we have answered an important question about enhancement of Stimulated Raman Scattering through control of the temporal behaviour of the pump and we are now in a position to consider some of the less well explored areas in this field.

5. Gain enhancement using a dual mode pump laser.

Some earlier authors have been concerned with the complicated four-wave mixing behaviour of the Stokes field when the pump field is a superposition of plane-wave modes with frequency separation much greater than the linewidth of the molecular oscillator Γ_A, (Stappaerts et al, 1980; Dzhotyan et al, 1977; Trutna et al, 1979; Eggleston et al, 1980; Ackerhalt, 1981). The motivation for this analysis may simply be that the pump laser output is unavoidably non-monochromatic, rather than theoretical support for an attempt to reduce the threshold pump power. However, we will discover that a real reduction in the pump power required to attain threshold is possible when the mode spacing is less than, or in the order of, the oscillator linewidth. The question we wish to answer in this section is: how does the Raman Gain depend on the distribution of pump energy amongst the closely spaced modes?

Berry and Hanna (1983), have described Stimulated Raman Scattering using a pump laser deliberately designed to deliver just two modes simultaneously. The characteristics of the Raman medium (which in this case is H_2 gas) can be adjusted to give a range of values for the oscillator bandwidth through pressure induced line broadening. In the following, we will attempt to quantify the effects of dual mode operation of the pump laser using the theory of section 4.

We are justified in using the result [28] for the Stokes intensity, but now the pump envelope is no longer constant in time:

$$|\epsilon_p|^2 = |\epsilon_1|^2 + |\epsilon_2|^2 + 2|\epsilon_1\epsilon_2|\cos(\Delta\omega.\tau+\phi) \qquad [40]$$

where $|\epsilon_1|$, $|\epsilon_2|$ are the mode amplitudes, and ϕ the relative phase (at $\tau = 0$). We will assume that the laser has been adjusted so that the modes have equal amplitude, whence

$$|\epsilon_p|^2 = \overline{|\epsilon_p|^2}[1 + \cos(\Delta\omega.\tau+\phi)] \qquad [41]$$

where the bar denotes a cycle average.

From this result it follows that:

$$\rho(\tau) - \rho(\tau') = \overline{|\epsilon_p|^2}\left[\, \tau - \tau' + \frac{2}{\Delta\omega}\, \sin\left(\frac{\Delta\omega}{2}(\tau-\tau')\right) . \cos\left(\frac{\Delta\omega}{2}(\tau+\tau') + \phi\right)\right] \qquad [42]$$

Long after the pump laser is switched on, the system will reach some 'dynamic equilibrium' :

$$I_s(z) = \underset{\tau\to\infty}{\mathrm{Lt}} \int_0^{\tau} d\tau' \left[\varepsilon_0 c\, \Gamma_A |\kappa_2|^2 / \rho_L\, \overline{|\epsilon_p|^2}\, z\right] . \left[1 + \cos(\Delta\omega\tau + \phi)\right]$$

$$. \exp\left[-2\Gamma_A(\tau-\tau')\right] . \left[I_0^2\left[\left(4\kappa_1\kappa_2 z(\rho(\tau)-\rho(\tau'))\right)^{1/2}\right] - I_1^2\left[\left(4\kappa_1\kappa_2 z(\rho(\tau)-\rho(\tau'))\right)^{1/2}\right]\right] \qquad [43]$$

where we have used [28] and [41]. Since the relative phase ϕ may take any value with equal probability, we may calculate the *expected* value of the Stokes intensity by averaging the result above:

$$I_s(z) = \underset{x\to\infty}{\mathrm{Lt}}\; I_{s0}^{(dm)}/_{2\pi} \int_0^{x} dx' \int_0^{2\pi} d\phi \;\left[\, 1 + \cos(\mu x + \phi)\right]$$

$$. \exp(-x') \left[I_0^2\left[\left(G_R z\left(x' + \frac{\sin \mu x'}{\mu} \cos\phi\right)\right)^{1/2}\right] - I_1^2\left[\left(G_R z\left(x' + \frac{\sin\mu x'}{\mu}\cos\phi\right)\right)^{1/2}\right]\right] \qquad [44]$$

where $\mu = \Delta\omega/(4\Gamma_A)$, $x' = 2\Gamma_A(\tau-\tau')$, and $I_{s0}^{(dm)} = 2G_R z\Gamma_A\tau_p I_{s0}^{(tr)}$ [45]

Also, the Raman gain is defined in terms of the (long) time averaged pump intensity:

$$G_R = 2k_1k_2\overline{|\epsilon_p|^2}/\Gamma_A \qquad [46]$$

The result [44] may be investigated under the the two limits of steady-state and transient response. The meaning of these is however modified from that of section 4 because we are not dealing with a top hat but rather a periodic envelope for the pump. Thus although the first term in [28] has been neglected, a 'transient' limit may be identified which leaves the Raman gain responding only to the *energy* in the pump over a period $2\pi/\Delta\omega$. I.e. it is sufficient that:

$$4\mu \gg 1; \quad G_R z \gg 1 \qquad [47]$$

in which case, from [44], we find that the averaged Stokes intensity is:

$$\langle I_s(z)\rangle \simeq I_{so}^{(ss)}\exp(G_R z) \qquad [48]$$

and the reader is reminded that the symbol G_R is proportional to the cycle-averaged pump intensity.

To find the steady-state limit of [44], it is simpler to assume that the classical steady-state result is valid, and thereby to qualify the result in terms of the constraints on the bandwidth. Thus we find that:

$$I_s(z,\tau;\phi) \simeq I_{so}^{(ss)}\exp[G_R z(1 + \cos(\Delta\omega.\tau+\phi))] \qquad [49]$$

which, with reference to [34], is seen to require:

$$1/(2\mu) > G_R z \gg 1 \qquad [50]$$

It follows from this constraint that the cycle-averaged (or phase-averaged) pump intensity is just:

$$\langle I_s(z)\rangle \simeq I_{so}^{(ss)}\exp(2G_R z) \qquad [51]$$

The results [51] and [48] mark the extremes for the averaged Stokes intensity for small and large values respectively of the ratio of pump to oscillator bandwidth. Our theory predicts a reduction in threshold pump intensity by a factor of ½ when the mode spacing is sufficiently small and we can measure this prediction against the results of Berry and Hanna as given in figure 1. If we assume that in this case, an exponential gain of about 30 was required in order that the (averaged) Stokes intensity attain (visible) threshold, then we might expect to see the minimum pump intensity required to occur for some value of $\Gamma_A/\Delta\omega > 15$. In fact we find from figure 1 that the reduction factor is about 0.6 for $\Gamma_A/\Delta\omega \simeq 2$ suggesting that the constraint [50] can be relaxed somewhat.

According to [47], the transient limit described by [48] can be trusted only for small values of $\Gamma_A/\Delta\omega$. Upon examination of figure 1, it seems plausable that the pump threshold power is then the same for both single and dual mode operation of the pump laser. The situation is complicated in this experiment however, by the finite overall duration of the pump pulse:

the steady-state condition [34] is violated when $\Gamma_A/\Delta\omega$ is very small, and the pump threshold *power* is therefore increased (see Berry and Hanna, 1983, for a fuller discussion).

6. Short pulses and spatial confinement

6.1 Introduction

We now devote our attention to a problem of great experimental interest, which involves an examination of the behaviour of a Stokes field when the active medium is excited by a short, pump pulse which is spatially 'confined' either by focussing, or by use of a guide. The aim is to obtain a result that quantifies the advantages of simultaneous spatial and temporal confinement in promoting Stimulated Raman Scattering.

Just as in chapter 2, it is convenient to partition the focussing problem by defining the two domains of high and low pump power. Unlike the work in that chapter however, it has not been possible to find a solution of the 'equations of motion' for the parameters of the Stokes beam that covers the general case for arbitrary pump power. However, useful asymptotic results valid in each of these domains have been obtained and there derivation is outlined below in sections 6.1 to 6.3. Section 6.4 brings together the results of chapter 3 with those of section 3 to give a desription of the evolution of a non-steady state Stokes field in a waveguide.

6.2 Low pump power focussed pump pulses

To be consistent with the approach of chapter 2, we would have to see the time dependent nature of the atom-field interaction, which manifests in section 2 of this chapter as equation [11], as contributing additional terms to the classical Lagrangian density. The procedure of that chapter could then be executed to recover the space and time dependent behaviour of the parameters of a Gaussian Stokes beam. This approach however has been unsuccessful; it has proved possible only to discover the solution in the extremes of high and low pump power. In this case, it is sufficient to retain the differential form of the Maxwell-Bloch equations for the problem, [11] and [15], to which the approximations suggested by these extremes may be applied directly.

We recall equations [15] and [11]:

$$\left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2ik_s\frac{\partial}{\partial z}\right]\epsilon_s = -2k_s K_2 q^* \epsilon_p \qquad [52]$$

$$\frac{\partial q}{\partial \tau} + \Gamma_A q = -iK_1 \epsilon_p \epsilon_s^* + F^* \qquad (\tau = t - z/c) \qquad [53]$$

where ϵ_p and ϵ_s are the slowly varying envelopes for the frequencies (ω_p, k_p) and (ω_s, k_s) respectively. We will now develop the low gain steady-state model of Boyd and Johnston (1969) by allowing ϵ_p and ϵ_s to vary in time. Their approach is simply to assume that the pump power is sufficiently small such that a first order perturbation theory suffices to describe the evolution of the free-space TEM_{nm} modes for the Stokes field. Concentrating on the TEM_{00} mode, we have:

$$\epsilon_s = \left[a_{00}(\tau, z)\, U_{00}(r, z; w_{s0}) + \sum_{(m,n)\neq(0,0)} a_{nm}(\tau, z)\, U_{nm}(r, z, \theta; w_{s0})\right] w_{s0}\sqrt{\frac{\pi}{2}} \qquad [54]$$

for which we will need the orthogonality condition:

$$\int_0^{2\pi}\int_0^{\infty} U_{00}(r, z; w_{s0})\, U_{nm}(r, z, \theta; w_{s0})\, r\, dr\, d\theta = \delta_{0,m}\,\delta_{0,n} \qquad [55]$$

where $U_{nm}(r,z,\theta;w_{s0})$ is the TEM_{nm} mode with focus coincident with that of the pump field, and which satisfies:

$$\left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2ik_s\frac{\partial}{\partial z}\right] U_{nm} = 0 \qquad [56]$$

The pump beam is also assumed to be a Gaussian beam, but with time varying envelope $\Omega(\tau)$:

$$\epsilon_p = \sqrt{\frac{\pi}{2}}\,\Omega(\tau)W_{po}U_{oo}(r,z;W_{po}) \qquad [57]$$

Our task now is to cast [52] into the recognisable form which is characteristic of time dependent plane wave Raman Scattering, and thereby employ the results of Raymer et al (1981) outlined in section 2. Combining [52] and [53] with [54] and [57], and making use of [55], we find to first order:

$$\frac{\partial a_{oo}}{\partial z} = -iK_2\,\breve{q}^{*}\,\Omega(\tau) \qquad [58]$$

$$\frac{\partial \breve{q}}{\partial \tau} + \Gamma_A\breve{q} = -iK_1\Omega^{*}(\tau)\,\omega^2(z)\,a_{oo} + \breve{F} \qquad [59]$$

where $$(\breve{q},\breve{F}) \equiv (W_{po}/W_{so})\int_0^{\infty}\int_0^{2\pi} U^{\dagger}_{oo}(r,z;W_{so})\,U_{oo}(r,z;W_{po})\,(q,F)\,d\theta\,r\,dr \qquad [60]$$

and $$\omega^2(z) = \frac{2}{\pi}\int_0^{2\pi}\int_0^{\infty} W_{po}^2\,|U_{oo}(r,z;W_{so})|^2\,|U_{oo}(r,z;W_{po})|^2\,r\,dr\,d\theta$$

$$= \frac{W_{po}^2}{W_p^2(z) + W_s^2(z)} \qquad [61]$$

It follows from the above definitions, and from [12] that:

$$\langle \breve{F}^{\dagger}(z,\tau)\,\breve{F}(z',\tau')\rangle = \frac{2\Gamma_A}{\rho_v}\,\omega^2(z)\,\delta(z-z')\,\delta(\tau-\tau') \qquad [62]$$

$$\langle \breve{q}^{\dagger}(z,0)\,\breve{q}(z',0)\rangle = \frac{1}{\rho_v}\,\omega^2(z)\,\delta(z-z') \qquad [63]$$

where ρ_v is the oscillator *volume* density. With the exception of the 'overlap' term $\omega^2(z)$, these equations are identical in form to those of section 2 in this chapter. In order to make the correspondence complete, it is convenient to make a change of variable:

$$\delta v_1/\delta z = \omega^2(z) \qquad [64]$$

$$(\hat{q},\hat{F}) = (\breve{q},\breve{F})/\omega^2(z) \qquad [65]$$

whence, the above becomes:

$$\langle \hat{F}^{+}(v_l,\tau)\, \hat{F}(v_l',\tau') \rangle = \frac{2\Gamma_A}{\rho_v}\, \delta(v_l - v_l')\, \delta(\tau-\tau') \qquad [66]$$

$$\langle \hat{q}^{+}(v_l,0)\, \hat{q}(v_l',0) \rangle = \frac{1}{\rho_v}\, \delta(v_l - v_l') \qquad [67]$$

and from [64] and [65], it now follows that:

$$\frac{\partial a_{00}}{\partial v_l} = -iK_2\, \hat{q}^{+}\, \Omega(\tau) \qquad [68]$$

$$\frac{\partial \hat{q}}{\partial \tau} + \Gamma_A \hat{q} = -iK_1 \Omega^{*}(\tau)\, a_{00} + \hat{F} \qquad [69]$$

The new independent variable v_l may be found from solving the equation:

$$\frac{dv_l}{dz} = \frac{W_{p0}^2}{W_p^2(z) + W_s^2(z)} \qquad [70]$$

for which we make the usual definitions:

$$\zeta = 2\left[\frac{z-f}{k_p W_{p0}^2}\right] \; ; \quad \mu = \frac{k_s W_{s0}^2}{k_p W_{p0}^2} \; ; \quad \kappa = \frac{k_s}{k_p} \qquad [71]$$

to obtain:

$$v_l = \frac{k_s W_{p0}^2}{2}\left[\kappa + \frac{1}{\mu}\right]^{-1/2}\left[\kappa+\mu\right]^{-1/2}\left[\tan^{-1}\left(\zeta\left[\frac{\kappa+\frac{1}{\mu}}{\kappa+\mu}\right]^{1/2}\right) - \tan^{-1}\left(\zeta_s\left[\frac{\kappa+\frac{1}{\mu}}{\kappa+\mu}\right]^{1/2}\right)\right] \qquad [72]$$

where $v_l(\zeta_s) = 0$. Equations [66-69] are now just the Maxwell-Bloch equations of section 3, so that the results of that section may be employed directly upon making the appropriate substitutions. Hence from [28]:

$$\langle |a_{00}(z,\tau)|^2 \rangle = 2\pi W_{p0}^2 \tfrac{1}{2}\varepsilon_0 c\, |\mathcal{E}(\tau)|^2\, v_l(z)\, |K_2|^2/\rho_l \; \cdot$$

$$\Big\{ e^{-2\Gamma_A\tau}\left[I_0^2\left[(4K_1K_2v_l\, p(\tau))^{1/2}\right] - I_1^2\left[(4K_1K_2v_l\, p(\tau))^{1/2}\right]\right]$$

$$+ 2\Gamma_A \int_0^{\tau} d\tau'\, e^{-2\Gamma_A(\tau-\tau')}\left[I_0^2\left[(4K_1K_2v_l(p(\tau)-p(\tau')))^{1/2}\right] - I_1^2\left[(4K_1K_2v_l(p(\tau)-p(\tau')))^{1/2}\right]\right]\Big\} \qquad [73]$$

where: $$p(\tau) = \int_0^{\tau} |\Omega(\tau'')|^2\, d\tau'' \qquad [74]$$

Thus we find, in the low gain approximation, the non-steady-state plane wave theory may be applied to the envelopes of the pump and Stokes Gaussian beams provided the z coordinate is replaced by $v_1(\zeta)$ of [72]. At this level of approximation, this substitution is therefore all that is required in order to utilise the family of curves published by Raymer and Mostowski. In the following section, we will analyse some predictions of this result and the necessary conditions for its validity.

6.4 Multipass gain-cell with short pump pulses

We now consider more specific experimental conditions under which we may employ [73]. Note that the steady-state limit of [73] is just the low-gain theory of Boyd and Johnston (1969) and Trutna and Byer (1981). However, if the multipass gain cell (see chapter 2) was driven by a top hat pump pulse with $\tau_p \ll 2\pi/\Gamma_A$, then we would be justified in taking the transient limit of [73]. This is a situation which has not yet been covered in the literature and is discussed in more detail below.

The slowly varying envelope of the Stokes intensity $\langle |a_{oo}(z,\tau)|^2 \rangle$, is a monotonic function of $v_1(\zeta)$, so that it is natural to assume that $v_1(\zeta)$ will be maximised (for fixed z) by appropriate choice of focussing conditions. It is easily seen from [72] that this requires:

$$\mu = 1; \qquad \text{i.e.} \qquad k_p w_{po}^2 = k_s w_{so}^2 \tag{75}$$

i.e. equal pump and Stokes confocal beam parameters. Under these optimal focussing conditions, $v_1(\zeta)$ becomes, for the first pass:

$$v_1(\zeta) = \tfrac{1}{2} k_s w_{po}^2 \theta(\zeta;\zeta_s)/(1 + \kappa) \tag{76}$$

where:

$$\theta(\zeta;\zeta_s) \equiv \tan^{-1}(\zeta) - \tan^{-1}(\zeta_s) \tag{77}$$

We will also assume that the resonator construction is such that the pump is tightly focussed (see the discussion in chapter 2, section 3.1), whereupon:

$$\theta(\zeta_e;\zeta_s) = \pi \tag{78}$$

For a multipass configuration, $v_1(\zeta)$ is the 'cumulative' distance traversed by the Stokes beam as it propagates around the cell and so, using [78] we define:

$$v_{1,n}(\zeta) = \tfrac{1}{2} k_s w_{po}^2 (n\pi + \delta\theta)/(1 + \kappa) \tag{79}$$

where:

$$\delta\theta = \{\ \theta(\zeta;\zeta_s)\ n \text{ even},\ \theta(\zeta;\zeta_e)\ n \text{ odd}\ \} \tag{80}$$

and n is the number of reflections suffered by the Stokes beam.

We will usually be justified in assuming the design of the system is such that the Stokes field has attained a threshold on leaving the cell. Therefore the total gain experienced by the Stokes field (from its initial value) will be large, and it will be safe to take the high gain transient limit of [73]:

$$\langle |a_{oo}(\omega,\tau_p)|^2\rangle \propto \exp\left[2\left[\frac{n\pi G_R k_s W_{po}^2 \Gamma_A \tau_p}{[1+\kappa]}\right]^{1/2}\right] \qquad [81]$$

where we have used [38] and [79]. Once again, we find that the (transient) exponential gain depends on the *energy* in the pump pulse but this time with the coordinate z replaced by the effective length $v_{1,n}$ given by [79]. Also it is interesting to note that the theory predicts a reduction by a factor $n^{-1/2}$ in pump energy required to pass the threshold.

Unfortunately, unlike the case discussed in chapter 2, the precise conditions defining the transient low gain domain are not known because we lack a unified (arbitrary gain) theory. However, we can argue, using the results of that chapter as a guide, for a set of approximate conditions necessary for the validity of [81]. These are summarised on the following page in table 4-1.

Table 4-1

Domain	Condition	Design constraints
transient	$\Gamma_A \tau_p << 1$	short pump pulse / long lifetime
tight focussing	$\zeta_e >> 1$; $\zeta_s << -1$	small waist / long cavity
low gain per pass	$2\left[\frac{\pi G_R K_s W_{po}^2 \Gamma_A \tau_p}{[1+K]}\right]^{1/2} \lesssim 1$	low pump pulse energy
high gain in total	$2\left[\frac{n\pi G_R K_s W_{po}^2 \Gamma_A \tau_p}{[1+K]}\right]^{1/2} >> 1$	many passes
optimal focussing	$K_s W_{so}^2 = k_p W_{po}^2$	equal confocal parameters

6.4 High power focussed pump pulses

A similar procedure to that above may be carried out to obtain a set of modified Maxwell-Bloch equations describing the evolution of the Stokes field in the high gain limit. Just as in chapter 2, we can expect the effects of gain-focussing to be such that, in the *high gain limit*, the Stokes waist is contained within that of the pump:

$$w_s(z)^2 \ll w_p(z)^2 \qquad [82]$$

Once again, we can use this assumption to justify a (radial) Taylor series expansion of the pump field retaining only terms upto quadratic. We begin with a substitution into [52] and [53] for the Stokes field:

$$\epsilon_s = a_{oo}(z,\tau)\exp[-iQ(z,\tau)r^2/2] \qquad [83]$$

and thereby obtain:

$$\left\{ \frac{\partial}{\partial\tau} + \Gamma_A - \frac{ir^2}{2}\frac{\partial Q}{\partial\tau} \right\}\left[q^* e^{iQr^2/2} \right] = iK_1 a_{oo}\epsilon_p^* + F e^{iQr^2/2} \qquad [84]$$

$$\left[Q^2 + K_s \frac{\partial Q}{\partial z} \right] \frac{r^2 a_{oo}}{2} + iQa_{oo} + iK_s\frac{\partial a_{oo}}{\partial z} = K_2 K_s q^* e^{iQr^2/2}\epsilon_p \qquad [85]$$

Consistent with the above, it is clear that we must expand not only the pump field, but also the oscillator coordinate and the Langevin function as a Taylor series:

$$q^* e^{iQr^2/2} = \sum_{n=0}^{\infty} b_n^+(z,\tau)\, r^{2n} \qquad [86]$$

$$F e^{iQr^2/2} = \sum_{n=0}^{\infty} f_n(z,\tau)\, r^{2n} \qquad [87]$$

$$\epsilon_p(r,z,\tau) = \sum_{n=0}^{\infty} c_n(z,\tau)\, r^{2n} \qquad [88]$$

so that substitution of [86], [87] and [88] into [84] and [85] gives a set of equations connecting the Stokes amplitude with the coefficients $b_n(z,\tau)$, $c_n(z,\tau)$, and $f_n(z,\tau)$.

Hence to zeroth order in r^2 we have:

$$\frac{\partial b_0^*}{\partial \tau} + \Gamma_A b_0^* = i K_1 a_{00} C_0^* + F_0 \qquad [89]$$

$$\frac{\partial}{\partial z} a_{00} + \frac{Q}{K_s} a_{00} = -i K_2 C_0 b_0^* \qquad [90]$$

whilst to first order:

$$\frac{\partial}{\partial \tau} b_1^* + \Gamma_A b_1^* = \frac{i b_0^*}{2} \frac{\partial Q}{\partial \tau} + i K_1 a_{00} C_1^* + F_1 \qquad [91]$$

$$\left[Q^2 + K_s \frac{\partial Q}{\partial z} \right] a_{00} = 2 K_2 K_s \left[b_0^* c_1 + b_1^* c_0 \right] \qquad [92]$$

Equations [89]-[92] describe the evolution of the Stokes field in the presence of a focussed, time-varying pump field. No satisfactory form for the analytic solution of these equations has been found. However we can make a few simple observations by comparing these with the standard Maxwell-Bloch equations for a plane wave pump. We notice that the equations for the zeroth order term are unchanged except for the 'diffraction loss' term involving the Stokes spot-size in [90]. Since, in chapter 2, it was demonstrated that the Stokes spot-size was a strong function of the pump intensity, we cannot ignore its variation with time through [92]. Phsically, there is an interaction between the time transients and the effects of gain focussing. However, just as for the high gain, tight focussing case discussed in chapter 2, we can conclude that the diffraction losses become relatively less important for sufficiently high gain, i.e. the very high gain limit of [89], [90] is just:

$$\frac{\partial \hat{b}_0^*}{\partial \tau} + \Gamma_A \hat{b}_0^* = i K_1 a_{00} \Omega^*(\tau) + \hat{F}_0 \qquad [93]$$

$$\frac{\partial a_{00}}{\partial v_h} \simeq -i K_2 \Omega(\tau) \hat{b}_0^* \qquad [94]$$

where we have used:

$$|C_0|^2 = |\Omega(\tau)|^2 \left[W_{p0} / W_p(z) \right]^2 \qquad [95]$$

$$V_h = \frac{k_p W_{p0}^2}{2} \Theta(\xi; \xi_s) \qquad [96]$$

and $$\left(\breve{b}_o^{+}, \breve{f}_o\right) = \left(b_o, f_o\right)\left[\frac{\Omega^*(\tau)}{C_o^*}\right]$$ [97]

giving $$\langle \breve{f}_o(\nu_h', \tau') \, \breve{f}_o^*(\nu_h, \tau) \rangle = \frac{2 P_A}{\rho_v} \, \delta(\nu_h - \nu_h') \, \delta(\tau - \tau')$$ [98]

Thus in this limit, the standard from is recovered with the change of variable [96] and so the results of the previous section [73] and [74], may thereby be adopted with the replacement $v_1 \rightarrow v_h$. Equally the high gain transient limit may be borrowed from that section, the exponential gain differing only by the factor $(K/(1 + K))^{1/2}$:

$$\langle |a_{oo}(z, \tau)|^2 \rangle \;\propto\; \exp\left[2 \left[\pi G_R \, K_p W_{po}^2 \, \Gamma_A \tau_p\right]^{1/2}\right]$$ [99]

Once again, the precise conditions which define the domain of very high gain are not known in the absence of a theory for arbitrary gain. However by analogy with the situation discussed in chapter 2, it is reasonable to assume that the theory is valid if the exponent in [99] is sufficiently large:

$$2 \left[\pi G_R \, K_p W_{po}^2 \, \Gamma_A \tau_p\right]^{1/2} \gg 1$$ [100]

As in the previous sction, we can argue for a set of approximate conditions necessary for the validity of [99], these are summarised in table 4-2.

Table 4-2

Domain	Condition	Design constraints
transient	$\Gamma_A \tau_p << 1$	short pump pulse / long lifetime
tight focussing	$\zeta_e >> 1$; $\zeta_s << -1$	small waist / long cavity
confined Stokes	$W_p^2(z) >> W_s^2(z)$	high pump pulse energy?
high gain in total	$2\left[\pi G_R K_p W_{po}^2 \Gamma_A \tau_p\right]^{1/2} >> 1$	high pump pulse energy

6.5 Transient scattering in a waveguide

Transient Stimulated Raman Scattering in a waveguide has recently been investigated as a means of dramatically reducing the pump power required to attain a threshold for the Stokes beam (Hanna et al, 1986). Indeed, one might expect a much reduced threshold power (assuming a continuous pump pulse train) as a result of combining the most effective means of spatial confinement with short pump pulses which take the scattering process into the transient regime. Unfortunately, a comparison of the results of Hanna et al with the predictions of the following model is not possible; the authors found that the (exponential) pump losses in the guide were much greater than that predicted by equation [42] of chapter 3. Their conclusion was that other loss mechanisms which have been excluded from our model (e.g. bending losses) were significant in their experiment.

In this section we demonstrate how the theory of section 2 of this chapter is readily applicable to a description of the scattering of short pump pulses in a capillary waveguide. The case for such a correspondence rests on the observation that the results of chapter 3 indicate that the Stokes profile remains unperturbed, except for the z dependent amplitude factor, as the field at that frequency propagates down the guide. Therefore we might expect to be able to modify the first order differential equations for the Stokes envelope in the presence of a plane-wave pump field (in free space) into a form consistent with the theory of propagation in a waveguide. The two factors to be accounted for by this modification are:

- The Stokes losses on reflection at the waveguide walls.

- The pump losses, and therefore the z dependent gain experienced by the Stokes field.

For a $LP_{\nu n}$ waveguide mode, each of these is properly accounted for by writing the Maxwell-Bloch equations ([19] and [20]) in the form:

$$\frac{\partial \epsilon_s(z,\tau)}{\partial z} = -\frac{\alpha_s}{2}\epsilon_s(z,\tau) - i k_2 \sqrt{C_{nn}^{\nu}}\, e^{-\alpha_p z/2}\, q^*(z,\tau)\, \epsilon_p(\tau) \qquad [101]$$

$$\frac{\partial q}{\partial \tau}(z,\tau) + \Gamma_A q(z,\tau) = -i\kappa_1^* \sqrt{C_{nn}^{v}}\; e^{-\alpha_p z/2}\, \epsilon_p(\tau)\, \epsilon_s^*(z,\tau) + F(z,\tau) \quad [102]$$

These equations can be solved upon making the appropriate transformations using the methods of section 3. First we define:

$$U = \epsilon_p^{-1}(0,\tau)\, \epsilon_s(z,\tau)\, \exp\left[\Gamma_A \tau + \alpha_s z/2\right] \quad [103]$$

and make the additional change of variable:

$$\sigma = \left[1 - e^{-\alpha_p z}\right] / \alpha_p \quad [104]$$

where σ is the new longitudinal co-ordinate (i.e. the effective length Z_{eff} of section 3). Using the above, equations [101] and [102] can be combined to give:

$$\frac{\partial^2 U}{\partial\sigma\partial\tau} - \kappa_1\kappa_2 |\epsilon_p(0,\tau)|^2 U = -i\kappa_2 F^* \exp\left[\Gamma_A \tau + (\alpha_s+\alpha_p)\frac{z}{2}\right] \quad [105]$$

which replaces equation [24] of section 3. With these substitutions, the only difference between [105] and [24] is in the source term which is responsible for initiating the scattering process. The full result for the Stokes field in a waveguide may therefore be written down on inspection:

$$\begin{aligned}\epsilon_s(z,\tau) = e^{-\alpha_s z/2}\Big\{ & \epsilon_s(0,\tau) - i\kappa_2 \epsilon_p(0,\tau) e^{-\Gamma_A \tau}\int_0^{\sigma} d\sigma'\, q^*(z',0)\, I_0\left[\left(4\kappa_1\kappa_2\, p(\tau)(\sigma-\sigma')\right)^{1/2}\right] \\ & + \sqrt{\kappa_1\kappa_2\sigma}\; \epsilon_p(0,\tau)\int_0^{\tau} d\tau' \left[p(\tau)-p(\tau')\right]^{-1/2} e^{-\Gamma_A(\tau-\tau')}\, \epsilon_p^*(0,\tau)\,\epsilon_s(0,\tau)\, I_1\left[\left(4\kappa_1\kappa_2(p(\tau)-p(\tau'))\sigma\right)^{1/2}\right] \\ & - i\kappa_2 \epsilon_p(0,\tau)\int_0^{\tau} d\tau' \int_0^{\sigma} d\sigma' \exp\left[-\Gamma_A(\tau-\tau') + (\alpha_p+\alpha_s)\frac{z'}{2}\right] F(z',\tau')\, I_0\left[\left(4\kappa_1\kappa_2(\sigma-\sigma')(p(\tau)-p(\tau'))\right)^{1/2}\right]\Big\}\end{aligned}$$

$$\text{where}\quad p(\tau) = C_{nn}^{v}\int_0^{\tau} |\epsilon_p(0,\tau'')|^2 d\tau'' \quad [106]$$

Equation [106] is valid for a Stokes field in a waveguide driven by a pump beam undergoing temporal variations of any kind. Unlike the free-space transient analysis of section 3 however, it is not possible to arrive at a simple expression for the Stokes intensity by forming the expectation $I_s = \frac{1}{2}\epsilon_0 c\langle E_s^* E_s\rangle$ and performing the integrals. This is because the 'natural' longitudinal co-ordinate for the pump ($\sigma = Z_{eff}$) is no longer that of the oscillator, and therefore of the noise source, as can be seen from equations [12]. However, we can be sure that the exact nature of the initiating process cannot effect the behaviour of the Stokes field once its magnitude has risen well above the noise. Note that the problem of conflicting 'natural co-ordinates' does not arise in the second term of [106] since this does not involve the Langevin function and the associated expectation value of equations [12]. Therefore it is convenient to invoke the fiction that the stimulated scattering process is driven by an injected Stokes field with an energy (density) much greater than that of the molecular oscillator in thermal equilibrium, whereupon all but the second term in [106] can be neglected (this is the approach taken by Carmen et al (1970)).

In this model then, [27] is no longer true, but instead the intensity of the Stokes field at the guide entrance is presumed whence a general expression for the intensity within the guide may be derived from [106]:

$$I_s(z,\tau) = \lim_{\tau \to \infty} I_{s0}\left[1 + e^{-\alpha_s z/2}\sqrt{k_1 k_2 \sigma}\,|\epsilon_p|^2 \int_0^{\tau} \frac{e^{-\Gamma_A(\tau-\tau')}}{[\tau-\tau']} I_1^2\left[\left(4 k_1 k_2 \sigma C^{v}_{nn} |\epsilon_p|^2 (\tau-\tau')\right)^{1/2}\right] d\tau'\right] \quad [107]$$

That this model gives the result for steady-state scattering of chapter 3 can be verified by extending the upper limit in the above to infinity ($\Gamma_a \tau_p \gg 1$) whereupon the integral can be performed to give (Gradshteyn and Ryzik):

$$I_{s,vn}(z) \simeq I^{(ss)}_{s,vn}(0)\exp[\,G_{vn}\sigma - \alpha_s z\,] \quad [108]$$

where $G_{vn} = 2C^{v}_{nn} k_1 k_2 |\epsilon_{po}|^2/\Gamma_A$ [109]

and $\sigma \equiv Z_{eff} = [1 - \exp(-\alpha_p z)]/\alpha_p$ [110]

which is just equation [62] of chapter 3.

Given then that [107] provides an accurate model of the Stokes intensity, we can now proceed to evaluate the integral in the limit of transient scattering. In this extreme, the integral is perfomed making use of the fact that the exponential is always nearly unity (see Carman et al, 1970), whereupon we obtain:

$$I_{s,\nu n}(z,\tau) \simeq I_{s,\nu n}(0)\exp[\ 2\left[2G_{\nu n}Z_{eff}\Gamma_A\tau\right]^{\frac{1}{2}} - \alpha_s z\ ] \qquad [111]$$

and this is valid for short pump pulses:

$$\Gamma_A\tau \ll 1 \qquad (\tau < \tau_p) \qquad [112]$$

and high gain:

$$2\left[2G_{\nu n}Z_{eff}\Gamma_A\tau\right]^{\frac{1}{2}} \gg 1 \qquad [113]$$

Borrowing from the analysis of section 3 in chapter 3, we find that the full Stokes field in a capillary waveguide can therefore be written:

$$E_s \simeq \Sigma_{\nu n}a_{\nu n}(0)J_{\nu n}(u_{\nu n}r/a)\exp[\ i\nu\theta + i\omega_s t - ik_s z + \tfrac{1}{2}\gamma_{\nu n}^{(tr)}\ ] \qquad [114]$$

where $$\gamma_{\nu n}^{(tr)} = 2\left[2G_{\nu n}Z_{eff}\Gamma_A\tau\right]^{\frac{1}{2}} - \alpha_s z \qquad [115]$$

is the net Stokes transient exponential gain (of the $LP_{\nu n}$ mode), and $a_{\nu n}(0)$ is the (supposed) injected amplitude of the $LP_{\nu n}$ mode polarised colinear with the pump field.

It is of interest to compare this result for transient scattering in a waveguide, with the result derived earlier in the chapter for plane-wave scattering in free-space. There are three changes that must be made to the latter in order to arrive at [114] (these are also the changes that should be made to convert the result for *steady-state* plane-wave Raman Scattering into that for *steady-state* Raman Scattering in a waveguide):

- The effective pump power (which appears in G, and $G_{\nu n}$ respectively) is modified by a factor C_{nn}^{ν}.

- The effective longitudinal co-ordinate is Z_{eff} which replaces z.

- The Stokes field suffers an additional exponential loss in the guide at a rate $\alpha_s z$

Just as for steady-state scattering in a waveguide, we find that the net transient Raman gain suffers from competition between the enhancing effects of guidance, and the depleting effects of losses at the waveguide walls. Once again, it is of interest to show how these effects depend on the design parameters available to the experimentalist. Therefore we define a set of normalised quantities which embody the available degrees of freedom (nominally: pump *energy*, waveguide bore, and waveguide length). Confining our attention to the LP_{01} mode, we let:

$$x_3 = k^2\gamma_{01}^{(tr)} \qquad [116]$$

which is the net exponential gain scaled by k^2,

$$x_2^3 = \alpha_p l \qquad [117]$$

which are the exponential pump losses, and

$$x_1 = 8k^4 G_{01}\Gamma_A\tau_p x_2/\alpha_p \qquad [118]$$

which is proportional to the energy in the pump pulse. Now [115] can be written:

$$x_3 = [1 - \exp(-x_2^3)]^{1/2}[x_1/x_2]^{1/2} - x_2^3 \qquad [119]$$

Once again the choice of these definitions is motivated by the need to decouple the dependencies of the $\{x_i\}$ on the intrinsic design variables, which are here taken to include guide bore and length. Unlike the steady-state case however, x_1 is proportional to energy in the pump pulse of duration τ_p (rather than pump power).

For the LP_{01} mode in a fused silica guide, with reference to [59], the $\{x_i\}$ may be written:

$$x_1 = 2.85(\lambda_p^{8}l/\lambda_s^{3})^{1/3}\ \hat{\mathcal{E}}_p \qquad [120]$$

$$x_2 = 0.75(\lambda_p^{2}l/a^{3})^{1/3} \qquad [121]$$

$$x_3 = \gamma_{01}^{(tr)}(\lambda_p^2/\lambda_s^2) \qquad [122]$$

where $\tilde{\mathcal{E}}_p$ is the normalised pump energy:

$$\tilde{\mathcal{E}}_p = \tilde{P}_p\Gamma_A\tau_p = (3/2)\,\mathrm{Im}\{\chi^{(R)}_{xx}\}k_s^2|\epsilon_{po}|^2 w_{po}^2\Gamma_A\tau_p \qquad [123]$$

The normalised variables x_2 versus x_1 are plotted in figure 2 for various values of x_3. Just as for the steady-state scattering in a waveguide, we find that the theory predicts an optimum design (in the sense that the pump energy required to attain threshold is minimised) for pump losses $x_2 \sim 1$: a guide narrower than this will give rise to increased pump losses, whilst a wider bore guide will fail to take full advantage of the opportunity for spatial confinement and therefore increased intensity. Since the qualitative nature of these curves are unchanged from those of figure 2 of chapter 3, then the optimisation procedures described in that chapter apply without qualification to figure 2 of this chapter and therefore are not repeated here.

In order to make a rough assessment of the advantages conferred by a capillary waveguide over free-space scattering, (both in the transient regime), we briefly compare the pump energy required to attain some fixed threshold when the guide bore radius is fixed. In this case the optimisation procedure may be shown to require:

$$x_3 \approx \sqrt{x_1/x_2} \qquad [124]$$

(see figure 2 for a confirmation of this relation). From [120-122], this in turn implies:

$$\gamma_{01}^{(tr)} \approx 2\left[\frac{\tilde{\mathcal{E}}_p a}{k\lambda_p}\right]^{1/2} \qquad [125]$$

for the net transient exponential gain. In terms of the normalised pump energy defined in [123], the transient exponential gain for a tightly focussed pump beam *in the high gain limit* from [99] is just:

$$\gamma^{(tr)} = 2\left[\frac{\tilde{\mathcal{E}}_p \pi}{k}\right]^{1/2} \qquad [126]$$

Comparison of [125] with [126] shows that, in this context, the use of a waveguide provides an increase in the exponential gain of the Stokes LP_{01}/TEM_{00} beam by a factor of approximately $\sqrt{a/\pi\lambda_p}$ for a fixed pump energy. Equivalently, this implies a reduction in pump energy required to attain threshold by a factor of $(a/\pi\lambda_p)$, and is unchanged from the result of section 4 in chapter 3.

7. Summary

In this chapter we have developed existing theory to cover a variety of (previously uninvestigated) cases in the field of Stimulated Raman Scattering. Whilst showing that gain enhancement results from pulse shortening (see the remarks at the end of section 4), we have also demonstrated the advantages of multimode operation of a c.w. pump laser in reducing the intensity required to attain threshold (section 5) for which the theoretical prediction is in close agreement with experimental results. In section 6 we have brought together results from earlier sections with those of chapters 2 and 3 to develop a theory covering simultaneously spatial and temporal confinement of the pump and Stokes beams. The success of this approach has depended on our ability to use the results appropriate to plane-wave Raman Scattering for the amplitude of the principle Stokes mode of the spatially confined field.

FIGURE 1

Pump threshold power versus Raman bandwidth - single and dual mode operation of pump laser.

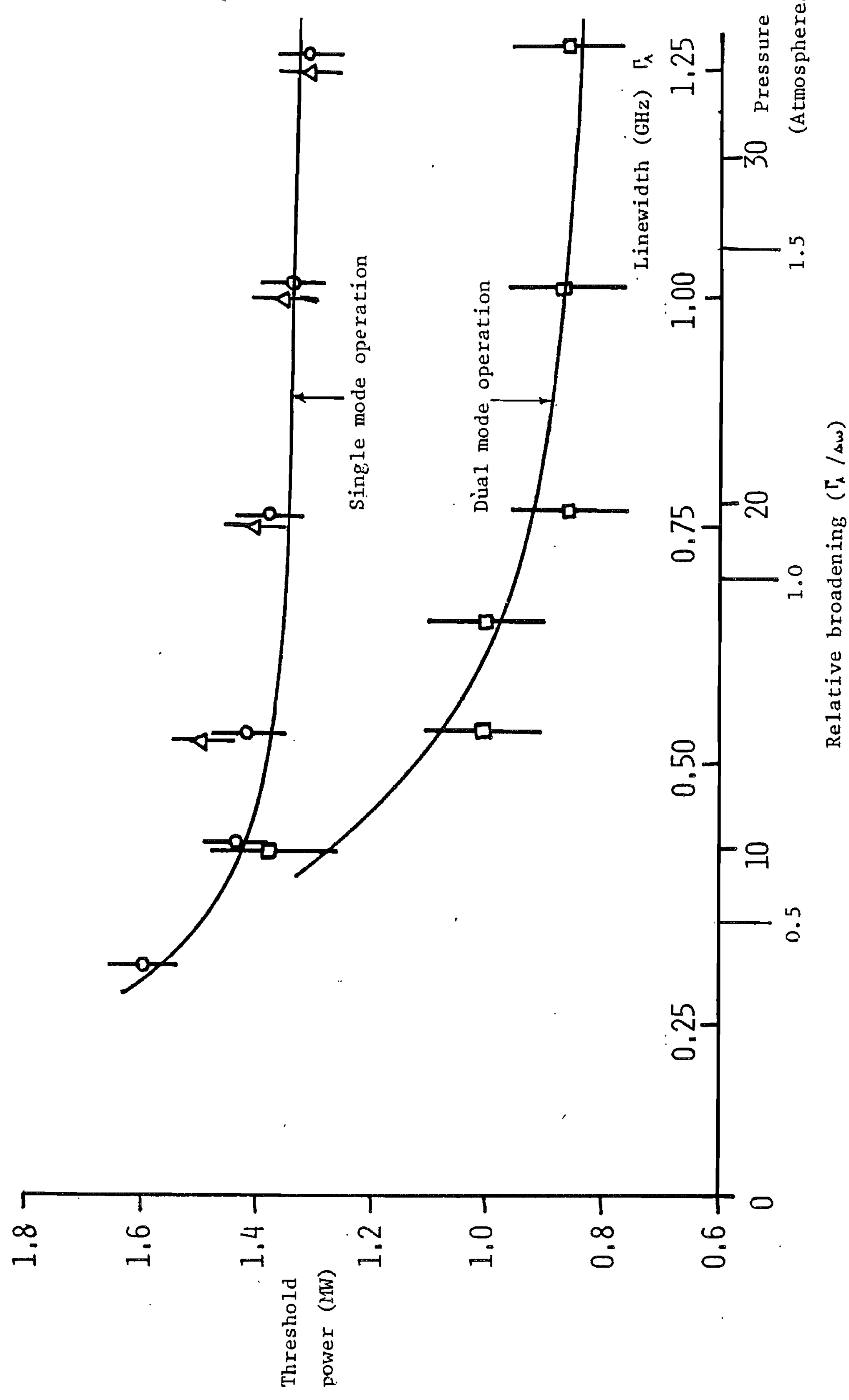
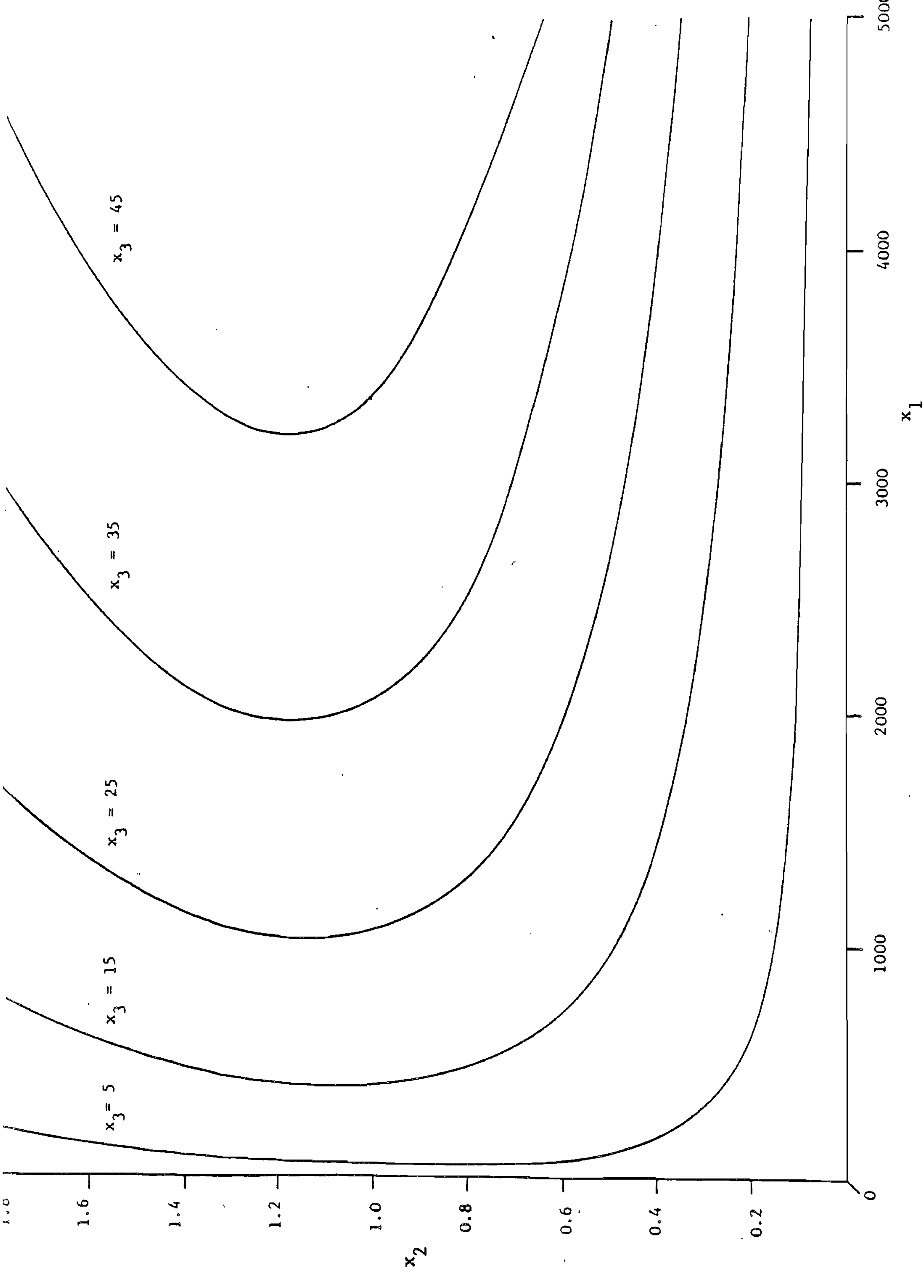


FIGURE 2

Curves of constant normalised gain (x_3) with normalised pump energy (x_1) and pump losses (x_2) for a Stokes beam in a Capillary Waveguide.



APPENDIX 1

SEPARABLE SOLUTIONS OF THE DIFFUSION EQUATION

1. Introduction

This appendix is devoted to an investigation of the scalar paraxial ray equation of chapter 1 which, in dimensionless co-ordinates may be written:

$$\{ \delta^2/\delta x^2 + \delta^2/\delta y^2 + \delta/\delta z + \beta(x,y,z) \} \, \phi(x,y,z) = 0 \qquad [1.1]$$

This investigation was initially prompted by two factors which arose in the work concerned with the spatial confinement problem of chapters 2 and 3. One of these was the need to find a co-ordinate transformation in which the paraxial ray equation associated with the gain-focussing problem of chapter 2 could by separated. The motivation was to pose this as an eigenvalue problem and thence to find a perturbative expansion for the eigenvalues. From this it would be a simple matter to integrate the remaining (separated) differential equation in the direction of propagation and so find the gain for the Stokes beam. Eventually this transformation was found (section 5), and work on a numerical solution to the eigenvalue problem was commenced. However the same discovery was made by Perry et al (1982), who had gone on to compute the numerical solution. At this point it was decided to seek a theoretical solution to the focussing problem since it was felt that such a solution, if discovered, would have some distinct advantages over that of a purely numerical treatment (see the discussion in chapter 2).

The second factor to influence this work arose from attepts to find an exact solution to the propagation problem for the Stokes beam in a waveguide (chapter 3). It was discovered that the perturbation expansion for the Stokes field in terms of Bessel functions (equaton [44] chapter 3) gave rise to a difference-differential equation in the expansion coefficients which could be transformed, through a suitable generating function, into a diffusion equation of the form:

$$\{ \delta^2/\delta x^2 + \delta/\delta z + \exp(-\alpha_p z)\cos(kx) \} \phi(x,z) = 0 \qquad [1.2]$$

Once again it was decided to try to find a transformation that would turn this into an eigenvalue problem making it susceptable to a perturbative treatment, and so the work in this direction which had been started on behalf of the focussing problem was expanded. Finally it was decided that a satisfactory conclusion would be attained only when an exhaustive search had been made for the full set of functions $\beta(x,y,z)$ for which [1.1] is separable. It was realised that the result of such an investigation would have some utility in other fields employing a diffusion equation with an inhomogeneous 'potential' $\beta(x,y,z)$, including thermodynamics and quantum mechanics.

The pupose of this appendix then, is to find the full set of potential functions $\beta(x,y,z)$ such that [1.1] separates into three ordinary differential equations in the appropriate transformed coordinate system. From this point (in two of the transformed coordinates) the theory of 2nd order differential equations will deliver exact or approximate solutions according to the complexity of terms within the allowed functional forms for $\beta(x,y,z)$. Thus it is possible, within this framework, to delineate all possible variations of refractive index that permit an exact solution to the scalar wave equation.

Section 2 describes the mechanics of the transfomation, whilst section 3 deals with the conditions for separability. Section 4 brings the results from these two sections together to give the set of separable transformations. Section 5 is a brief discussion of the implications of the result, both generally, and specifically with respect to some of the propagation problems analysed in this thesis.

2. Transformation of the diffusion equation

The general transformation of [1.1] is accomplished through a substitution of the form:

$$\{x,y,z\} \rightarrow \{u(x,y,z), v(x,y,z), w(x,y,z)\} \qquad [2.1]$$

and $$\phi(x,y,z) = \exp(Q(x,y,z))\psi(u,v,w) \qquad [2.2]$$

Here, u,v,w are the new coordinates spanning the transformed space. The exponential is an integrating factor providing a further degree of freedom which will be needed in the nest section. For the present, we will not discuss the formal properties of the mapping, but will tacitly assume that the inverse exists so that x,y,z and Q can happily be expressed in terms of the transormed coordinates (u,v,w).

Substitution of [2.1] and [2.2] into [1.1] yields:

$$\Big\{ (\underline{\nabla} u)^2 \frac{\partial^2}{\partial u^2} + (\underline{\nabla} v)^2 \frac{\partial^2}{\partial v^2} + (\underline{\nabla} w)^2 \frac{\partial^2}{\partial w^2} + 2\Big[\underline{\nabla} u \underline{\nabla} v \frac{\partial^2}{\partial u \partial v} + \underline{\nabla} v \underline{\nabla} w \frac{\partial^2}{\partial u \partial w} + \underline{\nabla} v \underline{\nabla} w \frac{\partial^2}{\partial v \partial w} \Big]$$

$$+ \nabla^2 \underline{u} + \frac{\partial \underline{u}}{\partial z} + 2(\underline{\nabla}^T Q)(\underline{\nabla}\, \underline{u}^T)\underline{\nabla}_u + \nabla^2 Q + (\underline{\nabla} Q)^2 + \frac{\partial Q}{\partial z} + \beta \Big\} \psi = 0 \qquad [2.3]$$

where
$$\underline{\nabla}^T \equiv \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, 0\right)$$

$$\underline{\nabla}_u \equiv \left(\frac{\partial}{\partial u}, \frac{\partial}{\partial v}, \frac{\partial}{\partial w}\right)$$

$$\underline{u}^T \equiv (u, v, w) \qquad [2.4]$$

We now demand that [2.4] represents a diffusion equation in the transformed space, and choose the propagation direction to be along the coordinate axis u=0, v=0. Then we can draw the folowing conclusions:

$$\underline{\nabla} u \underline{\nabla} w = 0$$

$$\underline{\nabla} v \underline{\nabla} w = 0$$

$$(\underline{\nabla} w)^2 = 0 \qquad [2.5]$$

Thus, the characteristics of w are seen to be everywhere orthogonal to those of u and v. provided that u and v are 'independent coordinates (i.e. the Jacobian J(u,v/x,y) does not vanish), then the conclusion must be that w is independent of x and y and so must be a function solely of z.

In the next section we wish to examine the conditions for separability of the transformed diffusion equation and therefore it is convenient to rewrite [2.3] as follows:

$$\left[a_1 \frac{\partial^2}{\partial u^2} + a_2 \frac{\partial^2}{\partial u \partial v} + a_3 \frac{\partial^2}{\partial v^2} + a_4 \frac{\partial}{\partial u} + a_5 \frac{\partial}{\partial v} + a_6 \frac{\partial}{\partial w} + a_7 \right] \varphi = 0 \qquad [2.6]$$

where

$$a_1 = (\underline{\nabla} u)^2$$

$$a_2 = 2 \underline{\nabla} u \underline{\nabla} v$$

$$a_3 = (\underline{\nabla} v)^2$$

$$a_4 = \nabla^2 u + \frac{\partial u}{\partial z} + 2 \underline{\nabla} Q \underline{\nabla} u$$

$$a_5 = \nabla^2 v + \frac{\partial v}{\partial z} + 2 \underline{\nabla} Q \underline{\nabla} v$$

$$a_6 = \frac{\partial w}{\partial z}$$

$$a_7 = \nabla^2 Q + (\underline{\nabla} Q)^2 + \frac{\partial Q}{\partial z} + \beta \qquad [2.7]$$

Hence, constraints on the functional forms of the $a_i(u,v,w)$ will furnish constraints on the behaviour of the functions $u(x,y,z)$, $v(x,y,z)$, $w(z)$ and $Q(x,y,z)$. The last of these will in turn constrain the form of $\beta(x,y,z)$ through the allowed forms for $a_7(u,v,w)$.

3. Conditions for separability

We will now consider, without reference to the result [2.7], which constraints exist on the functions $a_i(u,v,w)$ in order that [2.6] separates through a substitution

$$\psi = U(u)V(v)W(w) \qquad [3.1]$$

into 3 ordinary differential equations in the independent variables u,v,w. In the subsequent analysis we will make use of the following definitions:

- $f_i(u)$, $g_i(v)$, $h_i(w)$ are functions of a single variable;
- $p_i(u,v)$, $q_i(u,w)$, $r_i(v,w)$ are functions of a pair of variables;
- $s_i(u,v,w)$ is a general function of three variables.

Similarly, we will describe $\psi(u,v,w)$ in terms of the following functions:

- $U(u)$, $V(v)$, $W(w)$ (see [3.1] above);
- $X(u,v)$, $Y(u,w)$, $Z(v,w)$ are 'intermediate' functions.

We examine first the constraints on the $a_i(u,v,w)$ such that [3.2] separates in w first:

$$\psi = W(w)X(u,v) \qquad [3.2]$$

From [2.6] it follows that:

$$a_1\frac{X_{uu}}{X} + a_2\frac{X_{uv}}{X} + a_3\frac{X_{vv}}{X} + a_4\frac{X_u}{X} + a_5\frac{X_v}{X} + a_6\frac{W'}{W} + a_7 = 0 \qquad [3.3]$$

and for [3.3] to be separable this must be expressible as:

$$[p_a(u,v)h_a(w) + p_b(u,v)h_b(w)]s_a(u,v,w) = 0 \qquad [3.4]$$

for some set of functions $\{p_a,p_b,h_a,h_b,s_a\}$. Therefore [3.3] can be decomposed into two parts, each related to a term in [3.4] as follows:

$$a_1 X_{uu}/X + a_2 X_{uv}/X + a_3 X_{vv}/X + a_4 X_u/X + a_5 X_v/X = s_1 p_1 h_1 \quad [3.5]$$

$$a_6 W'/W = s_1 p_2 h_2 \quad [3.6]$$

$$a_7 = s_1 p_2 h_3 + s_1 p_3 h_1 \quad [3.7]$$

Examination of [3.5] to [3.7] reveals that the form [3.4] is preserved if we set:

$$s_a = s_1,\ h_a = h_1,\ h_b = h_2 + h_3,\ p_a = p_1 + p_3,\ p_b = p_2 \quad [3.8]$$

Thus, dividing by $s_1 p_2 h_1$, we have:

$$p_1/p_2 + p_3/p_2 = \lambda \quad [3.9]$$

$$h_2/h_1 + h_3/h_1 = -\lambda \quad [3.10]$$

where λ is the separation constant.

The constraints thus far on the $a_i(u,v,w)$ may be determined from equations [3.5–3.7]. Since the coefficients X_{uu}/X, X_{uv}/X etc. are themselves just functions of u and v, (and W_z/W is a function only of w), it is clear that the $a_i(u,v,w)$ must be expressible as follows:

$$a_1 = s_1 p_4 h_1$$

$$a_2 = s_1 p_5 h_1$$

$$a_3 = s_1 p_6 h_1$$

$$a_4 = s_1 p_7 h_1$$

$$a_5 = s_1 p_8 h_1$$

$$a_6 = s_1 p_2 h_4$$

$$a_7 = s_1 p_2 h_3 + s_1 p_3 h_1 \quad [3.11]$$

It follows that the separated equations must have the following form (combining [3.4-3.8]):

$$p_4 \frac{X_{uu}}{X} + p_5 \frac{X_{uv}}{X} + p_6 \frac{X_{vv}}{X} + p_7 \frac{X_u}{X} + p_8 \frac{X_v}{X} + p_3 - \lambda p_2 = 0 \qquad [3.12]$$

$$h_4 \frac{w'}{w} + h_3 + \lambda h_1 = 0 \qquad [3.13]$$

Note that [3.13] is an ordinary equation in w. This completes the first stage in the separation of [2.6] (in w first). The same procedure is used in the second stage to separate [3.12] into two ordinary differential equations in u and v. Just as before, we postulate a form:

$$[f_a(u)g_b(v) + f_b(u)g_b(v)]p_c(u,v) = 0 \qquad [3.14]$$

and decompose [3.12] accordingly:

$$p_4U''/U + p_7U'/U = f_1g_1p_9 \qquad [3.15]$$

$$p_6(U'/U).(V'/V) = 0 \qquad [3.16]$$

$$p_5V''/V + p_8V'/V = f_2g_2p_9 \qquad [3.17]$$

$$p_3 = [f_3g_1 + f_2g_3]p_9 \qquad [3.18]$$

$$p_2 = [f_4g_1 + f_2g_4]p_9 \qquad [3.19]$$

Examination of [3.15-3.19] reveals that the required form [3.14] is attained provided:

$$p_c = p_9,\ f_a = f_2,\ f_b = f_1 + f_3 + f_4,\ g_a = g_1 + g_3 + g_4,\ g_b = g_1 \qquad [3.20]$$

whence dividing by $f_2g_1p_9$ we obtain for some separation constant μ:

$$f_1/f_2 + f_3/f_2 - \lambda f_4/f_2 = \mu \qquad [3.21]$$

$$g_2/g_1 + g_3/g_1 - \lambda g_4/g_1 = -\mu \qquad [3.22]$$

The constraints on the $p_i(u,v)$ can now be found from equations [3.15-3.19]. Since the coefficients of U"/U and U'/U are themselves just functions of u, and coefficients V"/V and V'/V just functions of v, we must have:

$$p_2 = (f_4g_1 + f_2g_4)p_9$$

$$p_3 = (f_3g_1 + f_2g_3)p_9$$

$$p_4 = f_5g_1p_9$$

$$p_5 = 0$$

$$p_6 = f_2g_5p_9$$

$$p_7 = f_6g_1p_9$$

$$p_8 = f_2g_6p_9 \qquad [3.23]$$

Hence the (separated) ordinary differential equations are:

From [3.13]: $$h_4W' + (h_3 + \lambda h_1)W = 0 \qquad [3.24]$$

From [3.21]: $$f_5U'' + f_6U' + (f_3 - \lambda f_4 - \mu f_2)U = 0 \qquad [3.25]$$

From [3.22]: $$g_5V'' + g_6V' + (g_3 - \lambda g_4 + \mu g_1)V = 0 \qquad [3.26]$$

So far, no reference has been made to the transformation leading to the 'canonical form' [3.12] and [3.13]. With reference to the method of Section 2 of this appendix, we observe that the transformations x(u,v,w), y(u,v,w), z(u,v,w) and the 'integrating factor' exp[Q(u,v,w)] are all quite general. Therefore, we are free to assume (implicitly) a transformation on u : $u \to f(u)$ say, for any function f and also on the transformed variable U : $U \to F(U)$. This freedom can be exploited to eliminate unecessary (i.e. redundant) coefficients in equations [3.30-3.32]. One possibility is to set:

$$h_4 = f_5 = g_5 = 1 \qquad [3.33]$$

$$h_3 = f_6 = g_6 = 0 \qquad [3.34]$$

Using [3.11], [3.15], [3.33] and [3.34], the forms to be satisfied by the $a_i(u,v,w)$ are thus seen to be:

$$a_1 = s_2(u,v,w)g_1(v)h_1(w)$$

$$a_2 = 0$$

$$a_3 = s_2(u,v,w)f_2(u)h_1(w)$$

$$a_4 = 0$$

$$a_5 = 0$$

$$a_6 = s_2(u,v,w)[f_4(u)g_1(v) + f_2(u)g_4(v)]$$

$$a_7 = s_2(u,v,w)[f_3(u)g_1(v) + f_2(u)g_3(v)]h_1(w) \qquad [3.35]$$

and now the functions $U(u)$, $V(v)$ and $W(w)$ satisfy:

$$U'' + [f_3(u) - \lambda f_4(u) - \mu f_2(u)]U = 0 \qquad [3.36]$$

$$V'' + [g_3(v) - \lambda g_4(v) + \mu g_1(v)]V = 0 \qquad [3.37]$$

$$W' + \lambda h_1(w)W = 0 \qquad [3.38]$$

where we have defined:

$$s_2(u,v,w) = s_1(u,v,w)p_9(u,v) \qquad [3.39]$$

Equations [3.35-3.38] are the required results under the hypothesis that [1.1] separates in the variable w first. Thus we have a set of ordinary differential equations [3.36-3.38] in the functions $U(u)$, $V(v)$ and $W(w)$ with coefficients which are linear combinations of the functions $f_i(u)$, $g_i(v)$ and $h_i(w)$. These functions in turn constrain the functional behaviour of the coefficients $a_i(u,v,w)$ through [3.35].

If we require [1.1] to separate in the variable u first, i.e.:

$$\psi = U(u)Z(v,w) \qquad [3.40]$$

then the analysis of this section must be repeated in order to find the new constraints on the $a_i(u,v,w)$ and the corresponding new set of ordinary differential equations. The results are thus found to be:

$$a_1 = s_2(u,v,w)[g_4(v)h_1(w) + g_2(v)h_4(w)]$$

$$a_2 = 0$$

$$a_3 = s_2(u,v,w)f_1(u)h_1(w)$$

$$a_4 = 0$$

$$a_5 = 0$$

$$a_6 = s_2(u,v,w)f_1(u)g_2(v)$$

$$a_7 = s_2(u,v,w)\{f_3(u)[g_4(v)h_1(w) + g_2(v)h_4(w)] + f_1(u)g_3(v)h_1(w)\} \qquad [3.41]$$

for the $a_i(u,v,w)$, and:

$$U'' + [f_3(u) + \lambda f_1(u)]U = 0 \qquad [3.42]$$

$$V'' + [g_3(v) - \lambda g_4(v) - \mu g_2(v)]V = 0 \qquad [3.43]$$

$$W' + [\mu h_1(w) - \lambda h_4(w)]W = 0 \qquad [3.44]$$

are the ordinary differential equations in $U(u)$, $V(v)$ and $W(w)$. Note that *none* of the functions of [3.41-3.44] are related to those of [3.35-3.38].

Finally, we consider separation of the partial differential equation [1.1] in the variable v first. In this case we expect that ψ factorises as:

$$\psi = V(v)Y(u,w) \qquad [3.45]$$

However, from the symmetry of [1.1] and the generality of the transformation considered above, it is seen that this separation can offer no further degrees of freedom on the allowed forms for $a_i(u,v,w)$ once the functional relationship between the transformed variables (u,v,w) and the original variables (x,y,z) is made explicit. Therefore we will disregard this form for the function ψ and work with the results derived from the factorisations given in [3.2] and [3.40].

4. Solution of the Constraint Equations

In this section we embark on the solutions of the constraint equations for the $a_i(u,v,w)$ which combine the expressions for the transformation variables in [2.7] with the functional forms deduced in section 3 (i.e. those given in [3.35] and [3.41]). In section 4.1 we consider the allowed forms for the $a_i(u,v,w)$ given that ψ is separable in the variable w first. We find from these deliberations that there are two types of transformation each of which gives a distinct form for the $a_i(u,v,w)$. These are labelled as linear and non-linear transformations and are discussed respectively in Sections 4.1.1 and 4.1.2. In Section 4.2 we consider the allowed forms for the $a_i(u,v,w)$ given that ψ is separable in the variable u first. In this case we find that there is only one distinct form for the transformation and therefore for the $a_i(u,v,w)$.

4.1 Separation in w first

With reference to equations [3.35], it is convenient to make the substitution:

$$s_3(u,v,w) = s_2(u,v,w)f_2(u)g_1(v)h_1(w) \qquad [4.1]$$

whence [2,7] becomes:

$$(\nabla u)^2 = s_3(u,v,w) / f_2(u) \qquad [4.2]$$

$$\nabla u \nabla v = 0 \qquad [4.3]$$

$$(\nabla v)^2 = s_3(u,v,w)/g_1(v) \qquad [4.4]$$

$$\nabla^2 u + \frac{\partial u}{\partial z} + 2\nabla Q \nabla u = 0 \qquad [4.5]$$

$$\nabla^2 v + \frac{\partial v}{\partial z} + 2\nabla Q \nabla v = 0 \qquad [4.6]$$

$$\frac{\partial w}{\partial z} = s_2(u,v,w)\left[f_4(u)/f_2(u) + g_4(v)/g_1(v) \right] \qquad [4.7]$$

$$\nabla^2 Q + (\nabla Q)^2 + \frac{\partial Q}{\partial z} + \beta = s_3(u,v,w)\left[\frac{f_3(u)}{f_2(u)} + \frac{g_3(v)}{g_1(v)} \right] \qquad [4.8]$$

The first step is to obtain the functions $u(x,y,z)$, $v(x,y,z)$, and $w(z)$ from [4.2-4.4] and [4.7]. The next step will be to calculate a consistent Q from [4.5] and [4.6] whence finally β can be obtained from [4.8]. In order to achieve the first step, it is convenient to make some changes of variable:

$$q_1(u) = \int f_2^{1/2}(u)\, du \qquad [4.9]$$

$$q_2(v) = \int g_1^{1/2}(v)\, dv \qquad [4.10]$$

where it is to be understood that (ultimately) the q_i are given functions of u and v. The first step can now be expressed as the solution of the equations:

$$(\underline{\nabla} q_1)^2 = (\underline{\nabla} q_2)^2 = S_3\,(u,v,w) \qquad [4.11]$$

$$(\underline{\nabla} q_1)^2 = (\underline{\nabla} q_2)^2 = W_z\left[f_4(u)/f_2(u) + g_4(v)/g_1(v)\right]^{-1} \qquad [4.12]$$

and:
$$\underline{\nabla} q_1\, \underline{\nabla} q_2 = 0 \qquad [4.13]$$

Equations [4.11] and [4.13] yield the Cauchy-Riemann equations:

$$\frac{\partial q_1}{\partial x} = \frac{\partial q_2}{\partial y} \qquad [4.14]$$

$$\frac{\partial q_1}{\partial y} = -\frac{\partial q_2}{\partial x} \qquad [4.15]$$

for which the solution is easily found to be:

$$q_1 = F_1\,(x+iy, z) + F_2(x-iy, z) \qquad [4.16]$$

$$q_2 = \left[F_1\,(x+iy, z) - F_2(x-iy, z)\right]/i \qquad [4.17]$$

for any functions F_1 and F_2. Note that this is a conformal mapping from the space $\{x,y\}$ to $\{q_1,q_2\}$ only if $F_1 = F_2^*$. Note also that the transformations must yet be made to satisfy [4.12] which will give an additional constraint on the form of the q_i. From here onwards the dependence of the F_i on z will be taken for granted and the second argument thereof suppressed.

Through [4.9] and [4.10], we are at liberty to write the f_i as functions of q_1, and the g_i as functions of q_2. Therefore we will define:

$$G_1(q_1) = f_4(u)/f_2(u) \qquad [4.18]$$

$$G_2(q_2) = g_4(v)/g_1(v) \qquad [4.19]$$

where once again, the G_i are to be regarded as given functions. We will also find it convenient to make the change of variable:

$$\xi = x+iy\ ;\quad \xi^* = x-iy \qquad [4.20]$$

With these definitions, we are able to express the additional constraint on the functional behaviour of the q_i in [4.12] as follows:

$$4\, F_1'(\zeta)\, F_2'(\zeta^+) \left[G_1\left(F_1(\zeta) + F_2(\zeta^+)\right) + G_2\left(\frac{F_1(\zeta) - F_2(\zeta^+)}{i} \right) \right] = W_z \qquad [4.21]$$

Equation [4.21] can be integrated once it is recognised that the derivatives can be expressed as operators on the G_i:

$$\frac{\partial^2}{\partial\zeta\,\partial\zeta^+} \left[\iint G_1(t)\, dt^2 \Big|_{t = F_1 + F_2} + \iint G_2(t)\, dt^2 \Big|_{t = \frac{F_1 - F_2}{i}} \right] = \frac{W_z}{4} \qquad [4.22]$$

$$\Rightarrow \iint G_1(t)\, dt^2 \Big|_{t = F_1 + F_2} + \iint G_2(t)\, dt^2 \Big|_{t = \frac{F_1 - F_2}{i}} = \frac{W_z}{4} \left[F_3(\zeta) + F_4(\zeta^+) \right] \qquad [4.23]$$

and $F_3(\zeta)$, $F_4(\zeta^*)$ are constants of integration. Equation [4.23] can now be expressed as a purely functional equation. To make this apparent, we make the following definitions:

$$\alpha = F_1(\zeta) \quad ; \quad F_1^{-1}(\alpha) = H_1(\alpha)$$

$$\beta = F_2(\zeta^*) \quad ; \quad F_2^{-1}(\beta) = H_2(\beta) \qquad [4.24]$$

$$F_3(\zeta) = F_3(H_1(\alpha)) = I_1(\alpha)$$

$$F_4(\zeta^*) = F_4(H_2(\beta)) = I_2(\beta) \qquad [4.25]$$

$$(4/W_z) \iint G_1(t)dt^2|_{t=\alpha+\beta} = G_3(\alpha+\beta)$$

$$(4/W_z) \iint G_2(t)dt^2|_{t=(\alpha-\beta)/i} = G_4(\alpha-\beta) \qquad [4.26]$$

whereupon [4.23] becomes:

$$G_3(\alpha+\beta) + G_4(\alpha-\beta) = H_1(\alpha)H_2(\beta) + I_1(\alpha) + I_2(\beta) \qquad [4.27]$$

Perhaps surprisingly, given the generality of the analysis thus far, only one distinct possibility exists for the functions $\{G_3, G_4, H_1, H_2, I_1, I_2\}$. The proof is not given here, but the solution may be intimated from the work of Aczel, 1966 (after Wilson, 1926) on the equation [4.27] with

I_1, $I_2 \equiv 0$. In this case, the authors show that the functions G_i must be either the sum of two trignometric functions, or a quadratic function of the arguments ($(\alpha+\beta)$ and $(\alpha-\beta)$ respectively). In our case, it can be shown that both such solutions exist simultaneously in that the functions G_i may be a linear combination of trignometric *and* quadratic functions. It is instructive however, to construct two distinct solutions, which will be called linear and non-linear tranformations. The former is the solution to [4.27] with the trignometric components set to zero, whilst the latter is the full solution without any such constraints:

(i) Linear transformation

$$G_3(x) = p_3 + q_3x + r_3x^2$$

$$G_4(x) = p_4 + q_4x + r_4x^2$$

$$H_1(x) = \gamma_1 + b_1x$$

$$H_2(x) = \gamma_2 + b_2x$$

$$I_1(x) = p_1 + q_1x + r_1x^2$$

$$I_2(x) = p_2 + q_2x + r_2x^2$$

Substitution of the above into [4.27] gives the following constraints:

$$\gamma_1\gamma_2 + p_1 + p_2 = p_3 + p_4$$

$$b_1\gamma_2 + q_1 = q_3 + q_4$$

$$\gamma_1 b_2 + q_2 = q_3 - q_4$$

$$b_1 b_2 = 2(r_3 - r_4)$$

$$r_1 = r_2 = 2(r_3 + r_4) \qquad [4.28]$$

(ii) Non-linear transformation:

$$G_3(x) = p_3 + q_3x + r_3x^2 + t_3\cos(\omega x-\phi_3)$$

$$G_4(x) = p_4 + q_4x + r_4x^2 + t_4\cos(\omega x-\phi_4)$$

$$H_1(x) = \gamma_1 + b_1x + b_1\cos(\omega x-\theta_1)$$

$$H_2(x) = \gamma_2 + b_2x + b_2\cos(\omega x-\theta_2)$$

$$I_1(x) = p_1 + q_1x + r_1x^2 + t_1\cos(\omega x-\phi_1)$$

$$I_2(x) = p_2 + q_2x + r_2x^2 + t_2\cos(\omega x-\phi_2)$$

Substitution of the above into [4.27] gives the following constraints:

$$\gamma_1\gamma_2 + p_1 + p_2 = p_3 + p_4$$

$$b_1b_2 = 2t_3 = 2t_4$$

$$\gamma_1b_2 = t_2$$

$$b_1\gamma_2 = t_1$$

$$\theta_1 = \phi_1$$

$$\theta_2 = \phi_2$$

$$\theta_1 + \theta_2 = \phi_3$$

$$\theta_1 - \theta_2 = \phi_4$$

$$q_1 = q_3 + q_4$$

$$q_2 = q_3 - q_4$$

$$r_3 = r_4 = r_1/2 = r_2/2 \qquad [4.29]$$

Each of the coefficients (b_i, p_i, q_i, r_i, t_i, γ_i, θ_i, ϕ_i) in [4.28] and [4.29] is potentially a function of z. It is left to the analysis which follows to decide which of these must be constants, and which are true functional degrees of freedom. We note that the linear transformation [4.28] may be derived from the more general non-linear transformation [4.29] by defining a suitable limit for the coefficients as $\omega \to 0$. It will be seen that [4.28] gives rise to transformations which define Cartesian co-ordinates in the u,v plane, whilst the transformations resulting from [4.29] can give rise to cylindrical polar co-ordinates in the u,v plane. Each of these is treated separately in the following two sections.

4.1.1 The Linear Transformation

Equations [4.25] imply an inverse operation to recover the G_1, G_2:

$$G_1(\alpha+\beta) = (w_z/4)\left[\frac{\partial^2}{\partial t^2} G_3(\alpha+\beta)\Big|_{\alpha+\beta=t}\right]_{t=\alpha+\beta}$$

$$= w_z r_3/2 = A_1 \text{ say (from [4.28])} \qquad [4.30]$$

$$G_2((\alpha-\beta)/i) = (w_z/4)\left[\frac{\partial^2}{\partial t^2} G_4(\alpha-\beta)\Big|_{\alpha-\beta=it}\right]_{t=\frac{\alpha-\beta}{i}}$$

$$= -w_z r_4/2 = A_2 \text{ say (from [4.28])} \qquad [4.31]$$

From these we conclude that G_1, G_2 are independent of α and β and therefore of u and v. Therefore, from [4.18] and [4.19] we must have:

$$f_4(u) = A_1 f_2(u) \qquad [4.32]$$

$$g_4(v) = A_2 g_1(v) \qquad [4.33]$$

where we conclude from the definition of the G_i that the A_i cannot be functions of z. Also w_z can be expressed in terms of the $b_i(z)$ from [4.30] and [4.31] say (using [4.28]):

$$w_z = 4(A_1 + A_2)/(b_1(z)b_2(z)) \qquad [4.34]$$

and from [4.7] therefore we are able to deduce that the factor $s_3(u,v,w)$ must have the form:

$$s_3(u,v,w) = 4/(b_1(z)b_2(z)) \qquad [4.35]$$

The exact functional forms of the F_i can be recovered from [4.28] as follows:

$$F_1(\zeta) = (\zeta - \gamma_1(z))/b_1(z) \qquad [4.36]$$

$$F_2(\zeta^*) = (\zeta^* - \gamma_2(z))/b_2(z) \qquad [4.37]$$

so that from [4.17] we have the mapping of the functions $q_1(u)$, $q_2(v)$ into the x,y,z space:

$$q_1(u) \quad = \quad \frac{\zeta - \gamma_1(z)}{b_1(z)} + \frac{\zeta^+ - \gamma_2(z)}{b_2(z)} \qquad [4.38]$$

$$q_2(v) \quad = \quad \frac{\zeta - \gamma_1(z)}{i b_1(z)} - \frac{\zeta^+ - \gamma_2(z)}{i b_2(z)} \qquad [4.39]$$

Therefore the new co-ordinates (q_1,q_2) represent a linear transformation on the (x,y), whilst the (u,v) may have a more complex dependency through the functions $f_2(u)$, $g_1(v)$. Generally though, we can write:

$$u \quad = \quad q_1^{-1}\left(\frac{\zeta - \gamma_1}{b_1} + \frac{\zeta^+ - \gamma_2}{b_2} \right) \qquad [4.40]$$

$$v \quad = \quad q_2^{-1}\left(\frac{\zeta - \gamma_1}{i b_1} - \frac{\zeta^+ - \gamma_2}{i b_2} \right) \qquad [4.41]$$

Recalling [4.5] and [4.6], it remains only to find the Q consistent with these definitions for u and v. It is easily verified, using the complex co-ordinates $\zeta = x+iy$, $\zeta^* = x-iy$, and the results:

$$\nabla^2 q_i = 0 \quad ; \quad (\underline{\nabla} \cdot q_i)^2 = \frac{4}{b_1(z)\, b_2(z)} \qquad [4.42]$$

along with [4.38] and [4.39], that the equations in Q can be expressed in terms of the q_i as follows:

$$\nabla^2 u + \frac{\partial u}{\partial z} + 2\underline{\nabla} Q\, \underline{\nabla} v = f_2^{-1/2}\left[\frac{-2f_2'}{b_1 b_2 f_2^{3/2}} + \frac{\partial q_1}{\partial z} + \frac{4}{b_2}\frac{\partial Q}{\partial \zeta} + \frac{4}{b_1}\frac{\partial Q}{\partial \zeta^+} \right] \qquad [4.43]$$

$$\nabla^2 v + \frac{\partial v}{\partial z} + 2\underline{\nabla} Q\, \underline{\nabla} v = g_1^{-1/2}\left[\frac{-2g_1'}{b_1 b_2 g_1^{3/2}} + \frac{\partial q_2}{\partial z} - \frac{4}{i b_2}\frac{\partial Q}{\partial \zeta} + \frac{4}{i b_1}\frac{\partial Q}{\partial \zeta^+} \right] \qquad [4.44]$$

The 'active' variables in these equations are still (x,y,z), but for convenience, the functions $f_2(u)$ and $g_1(v)$ are left to imply a dependence on (x,y,z) through [4.40] and [4.39] repectively. It is now a simple matter to solve [4.43] and [4.44] for $\partial Q/\partial \zeta$ and $\partial Q/\partial \zeta^*$ giving two equations that must *simultaneously* be satisfied by the function Q(x,y,z):

$$\frac{\partial Q}{\partial \zeta} = \frac{1}{4b_1}\left[\frac{f_2'}{f_2^{3/2}} - \frac{i g_1'}{g_1^{3/2}} \right] - \frac{b_2}{8}\frac{\partial}{\partial z}(q_1 - iq_2) \qquad [4.45]$$

$$\frac{\partial Q}{\partial \zeta^+} = \frac{1}{4b_2}\left[\frac{f_2'}{f_2^{3/2}} + \frac{i g_1'}{g_1^{3/2}} \right] - \frac{b_1}{8}\frac{\partial}{\partial z}(q_1 + iq_2) \qquad [4.46]$$

Again, just as we have done earlier in this section, the integration is performed by first expressing the unknown functions in terms of derivatives using:

$$\frac{f_2'}{f_2^{3/2}} = b_1 \frac{\partial}{\partial \zeta} \ln f_2 = b_2 \frac{\partial}{\partial \zeta^+} \ln f_2 \qquad [4.47]$$

$$\frac{g_1'}{g_1^{3/2}} = i b_1 \frac{\partial}{\partial \zeta} \ln g_1 = -i b_2 \frac{\partial}{\partial \zeta^+} \ln g_1 \qquad [4.48]$$

where we have used [4.9], [4.10], [4.40] and [4.41]. Using these results, we can write the simultaneous equations, [4.45] and [4.46] in Q as:

$$\frac{\partial Q}{\partial \zeta} = \frac{1}{4} \frac{\partial}{\partial \zeta} \ln(f_2 g_1) + \frac{b_2}{4}\left[\zeta^+ \frac{b_2'}{b_2^2} + \left(\frac{\gamma_2}{b_2}\right)'\right] \qquad [4.49]$$

$$\frac{\partial Q}{\partial \zeta^+} = \frac{1}{4} \frac{\partial}{\partial \zeta^+} \ln(f_2 g_1) + \frac{b_1}{4}\left[\zeta \frac{b_1'}{b_1^2} + \left(\frac{\gamma_1}{b_1}\right)'\right] \qquad [4.50]$$

Upon integration of these equations, we find that Q must must satisfy:

$$Q = \frac{1}{4} \ln(f_2 g_1) + \frac{b_2}{4}\left[\zeta^+ \frac{b_2'}{b_2^2} + \left(\frac{\gamma_2}{b_2}\right)'\right]\zeta + k_3(\zeta^+, z) \qquad [4.51]$$

$$Q = \frac{1}{4} \ln(f_2 g_1) + \frac{b_1}{4}\left[\zeta \frac{b_1'}{b_1^2} + \left(\frac{\gamma_1}{b_1}\right)'\right]\zeta^* + k_4(\zeta, z) \qquad [4.52]$$

where we have introduced the functions $k_3(\zeta^*, z)$ and $k_4(\zeta, z)$ which are constants of integration. Since the above expressions for Q must be identical for all ζ, ζ^*, and z, it follows that the b_i and k_i must be related as follows:

$$b_2(z) = k_2 b_1(z) \qquad [4.53]$$

$$k_3(\zeta^*, z) = \frac{\zeta^+}{4} b_1 \left(\frac{\gamma_1}{b_1}\right)' + k_5(z) \qquad [4.54]$$

$$k_4(\zeta, z) = \frac{\zeta}{4} b_1 \left(\frac{\gamma_2}{b_1}\right)' + k_5(z) \qquad [4.55]$$

where k_2 is a constant, and $k_5(z)$ is an arbitrary function of z. Invoking these relationships, we find that the function Q has the form:

$$Q = \frac{1}{4} \ln(f_2 g_1) + \frac{\zeta \zeta^+ b_1'}{4 b_1} + \frac{\zeta}{4}\left[\gamma_2' - \frac{b_1'}{b_1}\gamma_2\right] + \frac{\zeta^+}{4}\left[\gamma_1' - \frac{b_1'}{b_1}\gamma_1\right] + k_5 \qquad [4.56]$$

The desired function β (equation [4.8]) can now be expressed as:

$$\beta(x,y,z) = \frac{4}{K_2 b_1^2(z)} \left[\frac{f_3(u)}{f_2(u)} + \frac{g_3(v)}{g_1(v)} \right] - \nabla^2 Q - (\underline{\nabla} Q)^2 - \frac{\partial Q}{\partial z} \qquad [4.57]$$

so that it is clear that we require the various derivatives of the result in [4.56]. The major steps to this calculation are as follows. First we note from the results of [4.9], [4.10], [4.40] and [4.41] that for any function H(u):

$$\frac{\partial}{\partial x} H(u) = f_2^{-1/2} H' \left[b_1^{-1} + b_2^{-1} \right] \qquad [4.58]$$

$$\frac{\partial}{\partial y} H(u) = i f_2^{-1/2} H' \left[b_1^{-1} - b_2^{-1} \right] \qquad [4.59]$$

Similarly for a function H(v):

$$\frac{\partial}{\partial x} H(v) = g_1^{-1/2} H' \left[b_1^{-1} - b_2^{-1} \right] \qquad [4.60]$$

$$\frac{\partial}{\partial y} H(v) = i g_1^{-1/2} H' \left[b_1^{-1} + b_2^{-1} \right] \qquad [4.61]$$

Making use of the above, the result of [4.56] and the relation [4.53] we obtain:

$$\nabla^2 Q = \frac{b_1'}{b_1} + \frac{1}{K_2 b_1^2} \left[\frac{f_2''}{f_2^2} - \frac{3}{2}\frac{f_2'^2}{f_2^3} + \frac{g_1''}{g_1^2} - \frac{3}{2}\frac{g_1'^2}{g_1^3} \right] \qquad [4.62]$$

A similar analysis yields the result for $|\underline{\nabla} Q|^2$:

$$(\underline{\nabla} Q)^2 = \frac{1}{4 K_2 b_1^2} \left[\frac{f_2'}{f_2^{3/2}} - \frac{i g_1'}{g_1^{3/2}} + \zeta^+ b_1' + b_1 \gamma_2' - b_1' \gamma_2 \right] \cdot \left[\frac{f_2'}{f_2^{3/2}} + \frac{i g_1'}{g_1^{3/2}} + K_2 \left(\zeta b_1' + b_1 \gamma_1' - b_1' \gamma_1 \right) \right] \qquad [4.63a]$$

Finally then, we evaluate the quantity $\partial Q/\partial z$:

$$\frac{\partial Q}{\partial z} = \frac{-1}{4 K_2 b_1^2} \Bigg\{ \frac{f_2'}{f_2^{3/2}} \left[K_2\left(\gamma_1' - \frac{b_1'}{b_1}\gamma_1\right) + \left(\gamma_2' - \frac{b_1'}{b_1}\gamma_2\right) + \frac{b_1'}{b_1}\Big(K_2(\zeta - \gamma_1) + (\zeta^+ - \gamma_2)\Big) \right]$$
$$- \frac{i g_1'}{g_1^{3/2}} \left[K_2\left(\gamma_1' - \frac{b_1'}{b_1}\gamma_1\right) - \left(\gamma_2' - \frac{b_1'}{b_1}\gamma_2\right) + \frac{b_1'}{b_1}\Big(K_2(\zeta - \gamma_1) - (\zeta^+ - \gamma_2)\Big) \right] \Bigg\}$$
$$+ K_5' + \frac{\zeta\zeta^+}{4}\left[\frac{b_1''}{b_1} - \left(\frac{b_1'}{b_1}\right)^2 \right] + \frac{\zeta}{4}\left[\gamma_2'' - \gamma_2\left[\frac{b_1''}{b_1} - \left(\frac{b_1'}{b_1}\right)^2\right]\right] + \frac{\zeta^+}{4}\left[\gamma_1'' - \gamma_1\left[\frac{b_1''}{b_1} - \left(\frac{b_1'}{b_1}\right)^2\right]\right] \qquad [4.63b]$$

and substitute this result along with [4.62] and [4.63] into the expression for β [4.57] to obtain:

$$\beta(x,y,z) = \frac{1}{k_2 b_1^2}\left[\frac{4f_3}{f_2} - \frac{f_2''}{f_2^2} + \frac{5}{4}\frac{f_2'^2}{f_2^3} + \frac{4g_3}{g_1} - \frac{g_1''}{g_1^2} + \frac{5}{4}\frac{g_1'^2}{g_1^3}\right] - \frac{b_1'}{b_1} - k_5'$$

$$- \frac{1}{4}\left[\zeta\zeta^{+}\frac{b_1''}{b_1} + \zeta\left(\gamma_2'' - \frac{b_1''\gamma_2}{b_1}\right) + \zeta^{+}\left(\gamma_1'' - \frac{b_1''\gamma_1}{b_1}\right) + \left(\gamma_1' - \frac{b_1'\gamma_1}{b_1}\right)\left(\gamma_2' - \frac{b_1'\gamma_2}{b_1}\right)\right] \quad [4.64]$$

This result expresses the full compliment of functional degrees of freedom for the function $\beta(x,y,z)$ given that the diffusion equation separates (according to the canonical form [2.6]) in w first with a 'linear transformation' expressed in the definitions [4.38] and [4.39].

Summary

It is perhaps useful at this point to review the definitions and relations supporting this result. First we will choose to regard the functions $f_i(u)$ and $g_i(v)$ as elementary (functional) degrees of freedom in u and v respectively; the functions $b_1(z)$, $\gamma_1(z)$, $\gamma_2(z)$ and $k_5(z)$ as elementary degrees of freedom in z; and k_2 as a constant degree of freedom. Then the co-ordinates u and v are defined in [4.40] and [4.41] through the intermediate functions q_i which are themselves defined in terms of the functions f_2 and g_1 in [4.38] and [4.39]. Making use then of the relation [4.53], the co-ordinate transformation for this case is:

$$u = q_1^{-1}\left(\frac{\zeta - \gamma_1}{b_1} + \frac{\zeta^{+} - \gamma_2}{k_2 b_1}\right) \quad [4.65]$$

$$v = q_2^{-1}\left(\frac{\zeta - \gamma_1}{i b_1} - \frac{\zeta^{+} - \gamma_2}{i k_2 b_1}\right) \quad [4.66]$$

(where q_1 and q_2 are defined in [4.38] and [4.39] respectively) and

$$w = \frac{8k_1}{k_2}\int \frac{1}{b_1^2}\,dz \quad [4.67]$$

with $$\zeta = x + iy \quad [4.68]$$

and $$\zeta^{+} = x - iy \quad [4.69]$$

Upon making this transformation, we find the solution of the diffusion equation can be written, recalling [2.2]:

$$\phi \quad = \quad \psi(u,v,w)\exp[Q(x,y,z)] \qquad [2.2]$$

where by virtue of its separability, the function $\psi(u,v,w)$ can be written:

$$\psi \quad = \quad \int d\mu \int d\lambda U(u;\mu,\lambda)V(v;\mu,\lambda)W(w;\lambda) \qquad [4.70]$$

where, recalling [3.36-3.38] and using [4.32] and [4.33], the functions U,V,W are solutions of the ordinary differential equations:

$$U'' \quad + \quad [f_3(u) \; - \; f_2(u)(\mu \; + \; \lambda A_1)]U \quad = \quad 0 \qquad [4.71]$$

$$V'' \quad + \quad [g_3(v) \; + \; g_1(v)(\mu \; - \; \lambda A_2)]V \quad = \quad 0 \qquad [4.72]$$

$$W' \quad + \quad [\lambda h_1(w)]W \quad = \quad 0 \qquad [4.73]$$

4.1.2 The Non-linear Transformation

Consider now the transformation given by equation [4.29] for which we proceed in an analagous manner the case discussed in section 4.1.1 above.

The functions G_1, G_2 are readily found from applying the operations depicted in [4.30] and [4.31] to the functions G_3 and G_4 respectively:

$$G_1(\alpha+\beta) = (w_z/8)[2r_1 - b_1 b_2 \omega^2 \cos(\omega(\alpha+\beta)-\phi_3)] \qquad [4.74]$$

$$G_2((\alpha-\beta)/i) = -(w_z/8)[2r_1 - b_1 b_2 \omega^2 \cos(\omega(\alpha-\beta)-\phi_4)] \qquad [4.75]$$

where α and β are unchanged from the definitions in [4.24].

Equations [4.18] and [4.19] imply that G_1, G_2 cannot be functions of z except through q_1, q_2 and therefore through α and β. Therefore the results [4.74] and [4.75] imply, through [4.27] that ϕ_3, ϕ_4 (and therefore ϕ_1, ϕ_2) and ω are constant whilst w_z and r_1 must satisfy:

$$r_1 = Bb_1(z)b_2(z)\omega^2/2$$

$$w_z = 8A/[\omega^2 b_1(z)b_2(z)] \qquad [4.76]$$

where A and B are constants. With these substitutions, the G_i are now:

$$G_1(q_1) = B - A\cos(\omega q_1-\phi_3) \qquad [4.77]$$

$$G_2(q_2) = A\cos(i\omega q_2-\phi_4) - B \qquad [4.78]$$

so that, recalling the relations [4.18] and [4.19], the above results imply:

$$f_4(u) = [B - A\cos(\omega q_1(u)-\phi_3)]f_2(u) \qquad [4.79]$$

$$g_4(v) = [A\cos(\omega q_2(v)-\phi_4) - B]g_1(v) \qquad [4.80]$$

Just as for the case discussed in section 4.4.1, we now recover the F_i from [4.29] and use this result to determine the intermediate transformed co-ordinates $q_1(u)$ and $q_2(v)$ according to the definitions in [4.16] and [4.17] whilst regarding the $b_i(z)$ as functional degrees of freedom:

$$\alpha = F_1(\zeta) = H_1^{-1}(\zeta) = [\phi_1 + \cos^{-1}((\zeta-\gamma_1(z))/b_1(z))]/\omega \quad [4.81]$$

$$\beta = F_2(\zeta^*) = H_2^{-1}(\zeta^*) = [\phi_2 + \cos^{-1}((\zeta^*-\gamma_2(z))/b_2(z))]/\omega \quad [4.82]$$

whereupon the q_i become:

$$q_1 = [\phi_3 + \cos^{-1}((\zeta-\gamma_1(z))/b_1(z)) + \cos^{-1}(\zeta^*-\gamma_2(z))/b_2(z))]/\omega \quad [4.83]$$

$$q_2 = [\phi_4 + \cos^{-1}((\zeta-\gamma_1(z))/b_1(z)) - \cos^{-1}(\zeta^*-\gamma_2(z))/b_2(z))]/i\omega \quad [4.84]$$

The new (non-linear) transformations then are:

$$u = q_1^{-1}\{[\phi_3 + \cos^{-1}((\zeta-\gamma_1(z))/b_1(z)) + \cos^{-1}(\zeta^*-\gamma_2(z))/b_2(z))]/\omega\} \quad [4.85]$$

$$v = q_2^{-1}\{[\phi_4 + \cos^{-1}((\zeta-\gamma_1(z))/b_1(z)) - \cos^{-1}(\zeta^*-\gamma_2(z))/b_2(z))]/i\omega\} \quad [4.86]$$

$$w = \int dz 8A/[\omega^2 b_1(z)b_2(z)] \quad [4.87]$$

where we recall that the functions q_i are given by [4.9] and [4.10]. The multiplicative function $s_3(u,v,w)$ is determined by [4.12]:

$$s_3(u,v,w) = w_z/(G_1 + G_2)$$

$$= 4\omega^{-2}[b_1^2 - (\zeta-\gamma_1)^2]^{-1/2}[b_2^2 - (\zeta^*-\gamma_2)^2]^{-1/2} \quad [4.88]$$

As before, we proceed with the solution of [4.5] and [4.6] for the integrating function Q. We first note the results:

$$\nabla^2 q_i = 0 \;, \qquad (\underline{\nabla} q_i)^2 = s_3(u,v,w) \qquad ; \; i = 1,2 \quad [4.89]$$

and apply the chain rule to express the derivatives of u and v in terms of q_1 and q_2 respectively whereupon equations [4.5] and [4.6] become:

$$\frac{f_2'}{2f_2^{3/2}} - \frac{\omega^2}{4}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{1/2}\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{1/2}\frac{\partial q_1}{\partial z}$$
$$+\ \omega\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{1/2}\frac{\partial Q}{\partial \zeta^+} + \omega\left[b_1^2-(\zeta-\gamma_1)^2\right]^{1/2}\frac{\partial Q}{\partial \zeta} = 0 \qquad [4.90]$$

$$\frac{ig_1'}{2g_1^{3/2}} - \frac{i\omega^2}{4}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{1/2}\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{1/2}\frac{\partial q_2}{\partial \zeta}$$
$$+\ \omega\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{1/2}\frac{\partial Q}{\partial \zeta^+} - \omega\left[b_1^2-(\zeta-\gamma_1)^2\right]^{1/2}\frac{\partial Q}{\partial \zeta} = 0 \qquad [4.91]$$

and therefore the function Q must satisfy:

$$\frac{\partial Q}{\partial \zeta} = \frac{-1}{4\omega}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{-1/2}\left[\frac{f_2'}{f_2^{3/2}} - \frac{ig_1'}{g_1^{3/2}}\right] + \frac{\omega}{8}\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{1/2}\frac{\partial}{\partial z}(q_1-iq_2) \qquad [4.92]$$

$$\frac{\partial Q}{\partial \zeta^+} = \frac{-1}{4\omega}\left[b_2^2-(\zeta^+-\gamma_2)^2\right]^{-1/2}\left[\frac{f_2'}{f_2^{3/2}} + \frac{ig_1'}{g_1^{3/2}}\right] + \frac{\omega}{8}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{1/2}\frac{\partial}{\partial z}(q_1+iq_2) \qquad [4.93]$$

Just as in the previous section, we proceed by expressing the derivatives of f_2, g_1 in a manner that prepares the ground for the integration of each of the two equations involving Q:

$$\frac{\partial Q}{\partial \zeta} = \frac{1}{4}\frac{\partial}{\partial \zeta}\ln(f_2 g_1) + \frac{b_2}{4}\left[\frac{b_2'}{b_2^2}\zeta^+ + \left(\frac{\gamma_2}{b_2}\right)'\right] \qquad [4.94]$$

$$\frac{\partial Q}{\partial \zeta^+} = \frac{1}{4}\frac{\partial}{\partial \zeta^+}\ln(f_2 g_1) + \frac{b_1}{4}\left[\frac{b_1'}{b_1^2}\zeta + \left(\frac{\gamma_1}{b_1}\right)'\right] \qquad [4.95]$$

These equations are now identical to [4.49] and [4.50], so that Q is clearly unchanged from that of the previous section:

$$Q = \frac{1}{4}\ln(g_1 f_2) + \frac{\zeta\zeta^+}{4}\frac{b_1'}{b_1} + \frac{\zeta}{4}\left[\gamma_2' - \frac{b_1'}{b_1}\gamma_2\right] + \frac{\zeta^+}{4}\left[\gamma_1' - \frac{b_1'}{b_1}\gamma_1\right] + k_5(z) \qquad [4.96]$$

where once again $b_2(z) = k_2 b_1(z)$ for some constant k_2. The desired function β in equation [4.8] can now be written:

$$\beta(x,y,z) = \frac{1}{\omega^2}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{-1/2}\left[b_2^2-(\zeta^+-\gamma_1)^2\right]^{-1/2}\left[\frac{4f_3}{f_2} + \frac{4g_3}{g_1}\right]$$
$$-\nabla^2 Q - (\underline{\nabla} Q)^2 - \frac{\partial Q}{\partial z} \qquad [4.97]$$

Clearly, the basic form of β remains unchanged from that of [4.64] and the final result can be written down upon inspection:

$$\beta(x,y,z) = \frac{1}{\omega^2}\left[b_1^2-(\zeta-\gamma_1)^2\right]^{-1/2}\left[b_1^2 K_2^2-(\zeta^+-\gamma_2)^2\right]^{-1/2}\left[\frac{4f_3}{f_2}-\frac{f_2''}{f_2^2}+\frac{5}{4}\frac{f_2'^2}{f_2^3}+\frac{4g_3}{g_1}-\frac{g_1''}{g_1^2}+\frac{5}{4}\frac{g_1'^2}{g_1^3}\right]$$

$$-K_5'-\frac{b_1'}{b_1}\,\frac{-1}{4}\left[\zeta\zeta^+\frac{b_1''}{b_1}+\zeta\left[\gamma_2''-\frac{b_1''\gamma_2}{b_1}\right]+\zeta^+\left[\gamma_1''-\frac{b_1''}{b_1}\gamma_1\right]+\left[\gamma_1'-\frac{b_1'\gamma_1}{b_1}\right]\left[\gamma_2'-\frac{b_1'}{b_1}\gamma_2\right]\right] \quad [4.98]$$

<u>Summary</u>

Just as for section 4.1.1, we recall the transformation necessary to convert [1.1] to a set of ordinary differential equations in U,V,W:

$$U = q_1^{-1}\left[\frac{1}{\omega}\left(\phi_3+\cos^{-1}\left(\frac{\zeta-\gamma_1}{b_1}\right)+\cos^{-1}\left(\frac{\zeta^+-\gamma_2}{K_2 b_1}\right)\right)\right] \quad [4.99]$$

$$V = q_2^{-1}\left[\frac{1}{i\omega}\left(\phi_4+\cos^{-1}\left(\frac{\zeta-\gamma_1}{b_1}\right)-\cos^{-1}\left(\frac{\zeta^+-\gamma_2}{K_2 b_1}\right)\right)\right] \quad [4.100]$$

$$W = \frac{8A}{K_2\omega^2}\int\frac{1}{b_1^2(z)}\,dz \quad [4.101]$$

where the q_i are given by [4.38] and [4.49]. Upon making this transformation and using [2.2]:

$$\phi = \psi(u,v,w)\exp[Q(x,y,z)] \quad [2.2]$$

where $$\psi = \int d\mu\int d\lambda\, U(u;\lambda,\mu)V(v;\lambda,\mu)W(w;\lambda) \quad [4.70]$$

and where, recalling [3.36-3.38] and using [4.79] and [4.80], the functions U,V,W must satisfy:

$$U'' + [f_3(u) + \{\lambda A\cos(\omega q_1(u)-\phi_3) - \lambda B - \mu\}f_2(u)]U = 0 \quad [4.102]$$

$$V'' + [g_3(v) + \{\lambda B - \lambda A\cos(\omega q_2(v)-\phi_4) + \mu\}g_1(v)]V = 0 \quad [4.103]$$

$$W' + \lambda h_1(w)W = 0 \quad [4.104]$$

4.2 Seperation in u first

We now consider the transformation that allows a separation in u first. With reference to equations [2.7] and [3.41] as before we make a further substitution:

$$s_3(u,v,w) = s_1(u,v,w)f_1(u)g_2(v)h_1(w) \qquad [4.105]$$

and the constraints on the transformation can thereby be written:

$$(\underline{\nabla} u)^2 = s_3(u,v,w)\left[g_4(v)/g_2(v) + h_4(w)/h_1(w) \right] / f_1(u) \qquad [4.106]$$

$$(\underline{\nabla} v)^2 = s_3(u,v,w) / g_2(v) \qquad [4.107]$$

$$\underline{\nabla} u \, \underline{\nabla} v = 0 \qquad [4.108]$$

$$\nabla^2 u + \frac{\partial u}{\partial z} + 2\underline{\nabla} Q \, \underline{\nabla} u = 0 \qquad [4.109]$$

$$\nabla^2 v + \frac{\partial v}{\partial z} + 2\underline{\nabla} Q \, \underline{\nabla} v = 0 \qquad [4.110]$$

$$\frac{dw}{dz} = s_3(u,v,w) / h_1(w) \qquad [4.111]$$

$$\nabla^2 Q + (\underline{\nabla} Q)^2 + \frac{\partial Q}{\partial z} + \beta = s_3(u,v,w)\left[\frac{g_3(v)}{g_2(v)} + \frac{f_3(u)}{f_1(u)}\left[\frac{g_4(v)}{g_2(v)} + \frac{h_4(w)}{h_1(w)} \right]\right] \qquad [4.112]$$

Equation [4.111] immediately gives that s_3 is a function of z only. We can therefore make the simplifying substitutions:

$$q_1(u) = \int f_1^{1/2}(u)\, du \qquad [4.113]$$

$$q_2(v) = \int g_2^{1/2}(v)\, dv \qquad [4.114]$$

whereupon [4.106-4.108] become:

$$(\underline{\nabla} q_1)^2 = s_3(u,v,w)\left[\frac{g_4(v)}{g_2(v)} + \frac{h_4(w)}{h_1(w)} \right] \qquad [4.115]$$

i.e. using s_3 is a function of z only, for some function $G(q_2,z)$ say:

$$(\underline{\nabla} q_1)^2 = G(q_2, z) \qquad [4.116]$$

and $$(\underline{\nabla} q_2)^2 = s_3(u, v, w) \qquad [4.117]$$

where $$\underline{\nabla} q_1 \cdot \underline{\nabla} q_2 = 0 \qquad [4.118]$$

From [4.117] we conclude that q_2 is a straight line in x,y space and from [4.118] that q_1 is everywhere in that space perpendicular to q_1. Therefore G can be a function of z only, and the q_i must have the form:

$$q_1 = \frac{\zeta - \gamma_1}{b_1} + \frac{\zeta^* - \gamma_2}{b_2} \qquad [4.119]$$

$$q_2 = \frac{\zeta - \gamma_1}{b_3} + \frac{\zeta^+ - \gamma_2}{b_4} \qquad [4.120]$$

where, by virtue of [4.118], the $b_i(z)$ must satisfy:

$$b_1(z)b_4(z) + b_2(z)b_3(z) = 0 \qquad [4.121]$$

and the $b_i(z)$ and $\gamma_i(z)$ are otherwise arbitrary functions of z. Furthermore, the constraint that G be a function of z only requires that:

$$g_4(v) = k_1 g_2(v) \qquad [4.122]$$

whilst [4.117] implies that:

$$s_3(u,v,w) = 4/[b_3(z)b_4(z)] \qquad [4.123]$$

whereupon [4.111] gives:

$$w_z = 4/[h_1(w)b_3(z)b_4(z)] \qquad [4.124]$$

Also [4.115] and [4.121] demand that:

$$h_4(w) + \{ k_1 + [b_3(z)/b_1(z)]^2 \}h_1(w) = 0 \qquad [4.125]$$

We now seek solutions of equations [4.109] and [4.110] for the function Q. The algebra proceeds much as in section 4.1, whereupon the modified equations are:

$$\nabla^2 u + \frac{\partial u}{\partial z} + 2\underline{\nabla} Q\, \underline{\nabla} u = f_1^{-1/2}\left[\frac{-2f_1'}{b_1 b_2 f_1^{3/2}} + \frac{\partial q_1}{\partial z} + \frac{4}{b_2}\frac{\partial Q}{\partial \zeta} + \frac{4}{b_1}\frac{\partial Q}{\partial \zeta^+}\right] \qquad [4.126]$$

$$\nabla^2 v + \frac{\partial v}{\partial z} + 2\underline{\nabla} Q\, \underline{\nabla} v = g_2^{-1/2}\left[\frac{-2g_2'}{b_3 b_4 g_2^{3/2}} + \frac{\partial q_2}{\partial z} + \frac{4}{b_4}\frac{\partial Q}{\partial \zeta} + \frac{4}{b_3}\frac{\partial Q}{\partial \zeta^+}\right] \qquad [4.127]$$

Therefore the function q must simultaneously satisfy:

$$\frac{\partial Q}{\partial \zeta} = \frac{1}{4}\left[\frac{f_1'}{b_1 f_1^{3/2}} + \frac{g_2'}{b_3 g_2^{3/2}}\right] - \frac{1}{8}\left[b_2\frac{\partial q_1}{\partial z} + b_4\frac{\partial q_2}{\partial z}\right] \qquad [4.128]$$

$$\frac{\partial Q}{\partial \zeta^*} = \frac{1}{4}\left[\frac{f_1'}{b_2 f_1^{3/2}} + \frac{g_2'}{b_4 g_2^{3/2}}\right] - \frac{1}{8}\left[b_1\frac{\partial q_1}{\partial z} + b_3\frac{\partial q_2}{\partial z}\right] \qquad [4.129]$$

Upon integration, and substitution of the results for q_i from [4.119] and [4.120], we find that the following condition must be satisfied by the $b_i(z)$:

$$b_1(z)b_3(z) = \text{constant}*b_2(z)b_4(z) \qquad [4.130]$$

Recalling [4.121], we find that the above implies:

$$b_2(z) = k_2 b_1(z) \qquad [4.131]$$

$$b_4(z) = -k_2 b_3(z) \qquad [4.132]$$

for some constant k_2, whence Q becomes:

$$Q = \frac{1}{4}\ln(f_1 g_2) + k_5 + \frac{\zeta\zeta^+}{8}\left[\frac{b_1'}{b_1} + \frac{b_3'}{b_3}\right] + \frac{1}{16}\left[k_2\zeta^2 + \frac{\zeta^{+2}}{k_2}\right]\left[\frac{b_1'}{b_1} - \frac{b_3'}{b_3}\right]$$
$$\frac{\zeta}{8}\left[k_2\left[b_1\left(\frac{\gamma_1}{b_1}\right)' - b_3\left(\frac{\gamma_1}{b_3}\right)'\right] + b_1\left(\frac{\gamma_2}{b_1}\right)' + b_3\left(\frac{\gamma_2}{b_3}\right)'\right] + \frac{\zeta^*}{8k_2}\left[k_2\left[b_1\left(\frac{\gamma_1}{b_1}\right)' + b_3\left(\frac{\gamma_1}{b_3}\right)'\right] + b_1\left(\frac{\gamma_2}{b_1}\right)' - b_3\left(\frac{\gamma_2}{b_3}\right)'\right] \qquad [4.133]$$

for some function $k_5(z)$.

Now the target function $\beta(x,y,z)$ from [4.112] can be written, using [4.121-4.123] and [4.125]:

$$\beta(x,y,z) = \frac{4}{k_2}\left[\frac{f_3}{f_1 b_1^2} - \frac{g_3}{g_2 b_3^2}\right] - \nabla^2 Q - (\underline{\nabla} Q)^2 - \frac{\partial Q}{\partial z} \qquad [4.134]$$

Upon evaluating the derivatives of Q in the normal way, after some lengthy manipulations, we obtain:

$$\begin{aligned}\beta(x,y,z) = {} & \frac{1}{k_2 b_1^2(z)}\left[\frac{-f_1''}{f_1^2} + \frac{5}{4}\frac{f_1'^2}{f_1^3} + \frac{4 f_3}{f_1}\right] - \frac{1}{k_2 b_3^2(z)}\left[\frac{-g_2''}{g_2^2} + \frac{5}{4}\frac{g_2'^2}{g_2^3} + \frac{4 g_3}{g_2}\right] \\ & + \frac{\zeta\zeta^+}{8}\left[\frac{b_1''}{b_1} + \frac{b_3''}{b_3}\right] - \frac{1}{16}\left[\frac{b_1''}{b_1} - \frac{b_3''}{b_3}\right]\left[k_2\zeta^2 + \zeta^{+2}/k_2\right] \\ & - \frac{\zeta}{4}\left[\gamma_2'' - \frac{\gamma_2}{2}\left[\frac{b_1''}{b_1} + \frac{b_3''}{b_3}\right] - \frac{k_2\gamma_1}{2}\left[\frac{b_1''}{b_1} - \frac{b_3''}{b_3}\right]\right] \\ & - \frac{\zeta^*}{4}\left[\gamma_1'' - \frac{\gamma_1}{2}\left[\frac{b_1''}{b_1} + \frac{b_3''}{b_3}\right] - \frac{\gamma_2}{2k_2}\left[\frac{b_1''}{b_1} - \frac{b_3''}{b_3}\right]\right] - \frac{1}{2}\left[\frac{b_1'}{b_1} + \frac{b_3'}{b_3}\right]\end{aligned}$$

[4.135]

Summary

Just as in section 4.1, we recall the transformations leading to this result, namely (from [4.119-4.121] and using [4.131] and [4.132]):

$$u = q_1^{-1}\left(\frac{\zeta - \gamma_1(z)}{b_1(z)} + \frac{\zeta^{t} - \gamma_2(z)}{k_2 b_1(z)} \right) \qquad [4.136]$$

$$v = q_2^{-1}\left(\frac{\zeta - \gamma_1(z)}{b_3(z)} - \frac{\zeta^{+} - \gamma_2(z)}{k_2 b_3(z)} \right) \qquad [4.137]$$

$$\int h_1(w)\, dw = -\frac{4}{k_2} \int \frac{1}{b_3^2(z)}\, dz \qquad [4.138]$$

where the q_i are defind in [4.113] and [4.114]. Upon making these substitutions, we find that the solution of the diffusion equation [1.1] can be written using [2.2]:

$$\phi(x,y,z) = \psi(x,y,z)\exp[\ Q(x,y,z)\ ] \qquad [2.2]$$

where $$\psi(x,y,z) = \int d\lambda \int d\mu\ U(u;\lambda)V(v;\lambda,\mu)W(w;\lambda,\mu) \qquad [4.139]$$

and, recalling [3.42-3.44], [4.122], [4.124] and [4.125] we find the functions U,V,W must satisfy the ordinary differential equations:

$$U'' + [f_3(u) + \lambda f_1(u)]U = 0 \qquad [4.140]$$

$$V'' + [g_3(v) - (\mu + k_1\lambda)g_2(v)]V = 0 \qquad [4.141]$$

$$W' + \{\mu - \lambda[k_1 + (b_3(z)/b_1(z))^2]\}h_1(w)W = 0 \qquad [4.142]$$

5. Conclusions

It may be seen from inspecting the results of sections 4.1 and 4.2 that the linear form of the transformation following a separation in u first is contained within the more general transformation which follows a separation in w first; thus by choosing $b_3(z)=ib_1(z)$ [4.135] becomes [4.64]. Therefore there are effectively two distinct types of transformation which render the diffusion equation [1.1] separable. Space does not permit a full discussion of the characteristics of each of the transformations although some of the features relevant to this work are briefly discussed below.

In the following section we discuss the results of this appendix with reference to the quadratic index profile $\beta \sim c(z)r^2$. In section 5.2 we show how the results of section 4.1.2 support the familiar transformation to a cylindrical co-ordinate system, and give the functional form for the index which permits separation. In section 5.3, this form is chosen to coincide with that arising from the gain-focussing problem of chapter 2 to give the eigenvalue problem of Perry et al.

5.1 Quadratic index in Cartesian co-ordinates

The relevance of this work to the problem of Raman Scattering follows from the identification of the paraxial ray equation (chapter 1) as a diffusion equation whence the potential function is then approximately proportional to the varying part of the refractive index. Particular forms for the transverse (x and y) variation of the refractive index have been used in the past to model the inhomogeneous properties of the medium and yet still permit a simple closed form solution for the propagating field. Almost universally, for a free-space, paraxial beam, these forms are based on a quadratic transverse variation of the refractive index. In particular, in their well known review, Kogelnik and Li (1966) showed how that, for a rotationally symmetric quadratic index variation, the field may be written as a series of modes which are Hermite-Gaussian functions in a Cartesian co-ordinate system. Shortly afterwards, Hanna (1969) pointed out that the Hermite-Gaussian beams were solutions to the paraxial wave equation when the refractive index variation was quadratic, but not rotationally invariant (i.e. astigmatic); a fact which has recently been rediscovered (Simon, 1985). A fuller, more detailed description of the types of field that result from a medium with a quadratic transverse index variation may be found in Arnaud (1970). With reference to the results of section 4.2 of this appendix, it can be shown that this type of solution arises (for instance) when the functions $f_3(u)$ and $g_3(v)$ are chosen to be quadratics, and the functions $f_1(u)$ and $g_2(v)$ are constant (see equation [4.135]). However, the advantage of our analysis is that the full compliment of functional degrees of freedom (that is variation in the direction of propagation) can be determined. Within this context then, considering only the mapping $(x,y) \rightarrow (u,v)$, we are able to interpret k_2 as the angular rotation, $(\gamma_1(z),\gamma_2(z))$ as the coordinate vector shift, and $b_1(z)^{-1}$, $b_2(z)^{-1}$ as the respective magnifications suffered by the axes in the transformation.

5.2 Cylindrical coordinates

It was mentioned in passing in section 4.1.2 that the transformation to a cylindrical co-ordinate system may be recovered from the more general transformation described in that section. Briefly, this may be achieved as follows. First we take the limit of [4.81] and [4.82] as:

$$\delta = e^{i\phi} \to 0 \text{ with } b_1(z) \to 2\delta s(z) \text{ and } A \to -2\delta^2 C \qquad [5.1]$$

for some arbitrary function s(z) and some constant C (note that these limits are consistent with the relations developed in section 4.2.1). Then, choosing:

$$B = 0,\ \gamma_1 = \gamma_2 = 0,\ k_2 = 1, \text{ and } \omega = -2i \qquad [5.2]$$

it is easily verified that the G_i become:

$$G_1 = Cr^2/s(z)^2 \qquad [5.3]$$

$$G_2 = 0 \qquad [5.4]$$

and that the transformation becomes:

$$q_1 = \ln(r/s(z)) \text{ where } r = (x^2 + y^2)^{1/2} \qquad [5.5]$$

and $$q_2 = \theta = \tan^{-1}(y/x) \qquad [5.6]$$

whereupon the potential function β given in [4.98] now has the form:

$$\beta = \frac{-1}{4r^2}\left[\frac{4f_3}{f_2} - \frac{f_2''}{f_2^2} + \frac{5}{4}\frac{f_2'^2}{f_2^3} + \frac{4g_3}{g_1} - \frac{g_1''}{g_1^2} + \frac{5}{4}\frac{g_1'^2}{g_1^3}\right] - \frac{r^2 s_1''}{4s_1} - \frac{s_1'}{s_1} - k_5' \qquad [5.7]$$

Equations [5.5] and [5.6] confirm the transfomation to a cylindrical co-ordinate system where, in this example, an arbitrary functional degree of freedom s(z) has been preserved.

5.3 Gain-focussing problem

As mentioned in the introduction to this appendix, the initial work on the separation problem was inspired by the gain-focussing problem which has been discussed in chapter 2. The relevance of the work of this appendix may be made apparent by first casting the paraxial ray equation ([12]) of that chapter into the canonical form [1.1]. This is achieved by equating β in [5.7] as follows:

$$\beta \;=\; i\widehat{P}_p \exp[-2r^2/w_p^2(z)]/w_p^2(z) \qquad [5.8]$$

We then choose in the above:

$$f_2(u) = u^{-2}; \quad g_1(v) = 1; \quad g_3(v) = 0; \quad C = 1 \qquad [5.9]$$

whereupon we have:

$$q_1(u) \;=\; \ln(u) \text{ so that } u \;=\; r/s(z);$$

Next we choose:

$$s(z) \;=\; w_p(z)$$

and replace z by $z/(-2ik_s)$ in [5.7], so that equating [5.7] and [5.8] leads to:

$$4\kappa^2u^2 + u^{-2}/4 - f_3(u) \;=\; i\widehat{P}_p \exp[-2u^2] \;\; ; \;\; (\kappa = k_s/k_p) \qquad [5.10]$$

and $\quad k_s' \;=\; -s'/s \qquad [5.11]$

The eigenvalue problem of interest comes from [4.102], which on substitution of [5.10] and using [5.3] becomes:

$$U'' \;+\; [(1/4-n^2)/u^2 + \lambda + 4\kappa^2u^2 - i\widehat{P}_p \exp(-2u^2)]U \;=\; 0 \qquad [5.12]$$

where, from the equation in the angular co-ordinate, we have deduced that $\mu \equiv n^2$ where n is an integer.

We have successfully transformed the paraxial ray equation for the gain-focussing problem discussed in chapter 2 into a set of ordinary differential equations. The above may now be treated as an eigenvalue problem, the solutions of which are required to vanish at infinity. This will provide a set of allowed values for λ which depend on the magnitude of the normalised pump power $\widehat{P}_p$, and thus determine the longitudinal gain through [4.104]. This was the method adopted by Perry et al (1982), whose approach was to expand U(u) as a sum of Gauss-Laguerre functions (Abromovitz and Stegun, 1970) and thereby treat the term due to the presence of the pump beam as a perturbation.

5.4 Summary

Clearly, the results of section 4 suggest that a variety of possibly previously undiscovered refractive index variations exist for which an exact separation of the paraxial ray equation is possible. In addition, because this has been an exhaustive search, the results of that section may be useful in showing that for some particular forms for the refractive index, no transformation exists for which the system is separable. Indeed, we can see at a glance that no separation is possible under any transformation for the potential function in [1.2].

Note however that not all forms for the refractive index which give rise to separable transformations will also give rise to ordinary differential equations that can be solved in terms of tabulated functions. Indeed, the work of Perry et al is an illustration of this point. Further work remains to be done therefore, in order to isolate the functional forms for the refractive index that permit an exact solution in the separated co-odinate system in terms of tabulated functions. One possibility is to closely follow the work of Westcott (1968a, 1968b) and insist that the potential terms involving the functions $f_i(u)$, $g_i(v)$ in the transformed system are such as to allow a solution in terms of hypergeometric functions. This constraint could then be 'propagated backwards' to identify the constraints on the refractive index variation. It is hoped that this possibility will be investigated in the near future.

APPENDIX 2

PAPER: ANALYSIS OF RAMAN GAIN FOR FOCUSSED GAUSSIAN BEAMS

Preface

The following paper has been submitted to Applied Physics B under the above title. The contents closely follow the exposition of chapter 2 in the main body of the thesis.

Title: **Analysis of Raman Gain for Focussed Gaussian Pump Beams**

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Abstract

Several theoretical and numerical models have been published which describe the evolution of a Stokes beam in a Raman medium excited by a focussed pump beam. Generally, the published theoretical departures from the plane-wave theory of Raman Scattering are based on assumptions about the power of the pump beam. In this paper we present a theoretical model which is shown to be in excellent agreement with an exact numerical treatment, and which is valid without restrictions on the pump power. Its predictions are used to indicate the range of validity of earlier theories.

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1. Introduction

Several authors have addressed the problem of describing the spatial behaviour of a Stokes field in a Raman active medium driven by a focussed pump beam [1-7]. The motivation is generally to discover the optimum experimental conditions required to effect an efficient conversion of energy from the pump to the Stokes field. The information required may include the necessary pump power such that the Stokes field attain some threshold, the optimum focussing conditons for the pump beam that minimise the threshold, and the Stokes beam parameters on leaving the medium.

In this paper, we present an approximate solution to the problem using a variational technique for describing the evolution of the Stokes field in the gain medium as a Gaussian (TEM_{00}) beam. This solution is shown to be in excellent agreement with an exact numerical treatment of Perry et al [5,6], and that the work of earlier authors [1-4] are special cases of the general results we derive, each having associated limited domains of applicability. Thus the 'overlap integral' approach of Boyd et al [1] is seen to be the limit for low pump power in our more general result, whilst the solution developed by Cotter et al [2] based upon a quadratic index profile approximation is shown to be applicable only when the pump power is sufficiently large.

The following section deals with the derivation of the equations of motion for the parameters describing the Stokes field. The next section presents the solutions to these equations under the high and low pump power limits discussed above followed by the more general result of our analysis. The final section is a discussion of the predictions of this result together with a comparison with the results of Perry et al [5,6].

2. Equations of motion for the Stokes field

In the following analysis we assume that the Stokes field growth is small signal, steady-state, and without competing processes. The conditions to be satisfied are respectively:

(a) The intensity of the Stokes field is not large enough to deplete the pump or saturate the medium.
(b) The pump and Stokes field each have a bandwidth smaller than the Raman linewidth [8].
(c) The gain and material dispersion of the medium favours the dominant growth of a field at the first Stokes frequency over higher order Raman processes [9].

Our starting point in the variational approach to the derivation of the Stokes field is the Lagrangian density for the electromagnetic field [10]:

$$\mathcal{L} = \frac{1}{2}\left[\underline{D}\cdot\underline{E} - \underline{B}\cdot\underline{H}\right] \tag{1}$$

and the Maxwell relation:

$$\frac{\partial \underline{H}}{\partial t} = \frac{1}{\mu_0}\,\underline{\nabla}\times\underline{E} \tag{2}$$

The pump and Stokes fields are defined as those components of the total field with frequencies ω_p and ω_s respectively. In the small signal regime, the pump field is unpertubed by the medium and its spatial distribution may therefore be regarded as given. Thus (1) and (2) apply to the field components at the Stokes frequency only, which we expand in the usual manner making explicit the rapidly varying part of the spatial variation in the z direction:

$$\underline{E} = \mathrm{Re}\left\{ \epsilon_s(\underline{r}) \exp\left[i\left(\omega_s t - k_s z\right) \right] \right\} \hat{\underline{e}}_s \tag{3}$$

and similarly for the magnetic field. Here $\hat{\underline{e}}_s$ is a unit vector in the direction of polarisation of the Stokes field, and $k_s = n_s\omega_s/c$, n_s is the refractive index at the Stokes frequency, and $\epsilon_s(\underline{r})$ is a slowly varying envelope. In addition to (a), (b), and (c), it is assumed in this paper that the pump field is a Gaussian beam and that the Stokes field is linearly polarised parallel to the pump field. Then the fields $\underline{D}$ and $\underline{B}$ can be written in terms of the electric and magnetic field vectors as follows:

$$\underline{B} = \mu_0 \underline{H} \quad ; \quad \underline{D} = \varepsilon_0 n_s^2 \underline{E} + \underline{P} \tag{4}$$

$$\underline{P} = \tfrac{3}{2}\, \varepsilon_0 \left|\epsilon_p(\underline{r})\right|^2 \chi^{(R)} \underline{E} \tag{5}$$

$$\left|\epsilon_p(\underline{r})\right|^2 = \left|\epsilon_{p0}\right|^2 \left[\frac{W_{p0}}{W_p(z)}\right]^2 \exp\left[-\frac{2r^2}{W_p^2(z)} \right] \tag{6}$$

$$W_p^2(z) = W_{p0}^2 \left[1 + 4\left[\frac{z-f}{k_p W_{p0}^2}\right]^2 \right] \tag{7}$$

where $\chi^{(R)}$ is the Raman susceptibility, the definition of which is taken from Hanna et al [11]. Classically, the Stokes field $\epsilon_s(\underline{r})$ will be that distribution for which the integral of the Lagrangian density is a minimum:

$$\delta \left\{ \iiiint \mathcal{L}\left(\underline{E}, \underline{H}\right) dx\, dy\, dz\, dt \right\} = 0 \tag{8}$$

We now make use of the paraxial approximation:

$$\left|\frac{\partial \epsilon_s}{\partial z}\right| \ll k_s \left|\epsilon_s\right| \tag{9}$$

and the boundary conditions:

$$\mathrm{Lt}_{x,y \to \infty}\, \epsilon_s(\underline{r}) = 0 \; ; \qquad \epsilon_s(\underline{r})\Big|_{z=0} \text{ given} \tag{10}$$

so that the Lagrangian density becomes, upon substitution of (2)-(7) into (1):

$$\mathcal{L} = \frac{1}{4\mu_0\omega_s^2}\left[\frac{3}{2}\frac{k_s^2}{n_s^2}\chi^{(R)}\left|\epsilon_p\epsilon_s\right|^2 + 2\,\mathrm{Im}\left\{k_s\epsilon_s^*\frac{\partial\epsilon_s}{\partial z}\right\} - \left|\frac{\partial\epsilon_s}{\partial x}\right|^2 - \left|\frac{\partial\epsilon_s}{\partial y}\right|^2\right] \tag{11}$$

Here it is assumed that the integrals over z and t of the Lagarangian density extend over many cycles of the Stokes field. Therefore the rapidly varying components in z and t do not contribute to (1) and have been omitted from (11). The Euler-Lagrange equation for the above is just the paraxial ray equation:

$$\left\{\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - 2ik_s\frac{\partial}{\partial z} + \frac{3}{2}\frac{k_s^2}{n_s^2}\chi^{(R)}\left|\epsilon_p\right|^2\right\}\epsilon_s = 0 \tag{12}$$

a full solution of which has been sought by Perry et al [5,6], and more recently by Gavrielides and Peterson [7] who have also taken into account depletion of the pump beam. Their approach was to pose (12) as an eigenvector problem in the Hilbert space of Gauss-Laguerre functions which are the TEM freespace modes. The associated eigenvalues represent the growth of the Stokes beam on propagation through the gain medium. For the particular case $k_s = k_p$, Perry et al give their results for the variation of the three largest eigenvalues with the pump power. Although theirs is an exact (numerical) solution of (12), an approximate analytic treatment would in some cases be more desirable. For instance, one is generally interested in the component of the Stokes beam that couples into an optimally chosen TEM_{00} beam, whereas the spatial transverse profile of the Stokes beam at the exit of the gain medium is not readily recoverable from the Gauss-Laguerre eigenvectors.

The following treatment therefore models the Stokes field as a Gaussian beam throughout the medium, the parameters of which are chosen to minimise (11). Our approximation consists of ignoring the coupling between this and higher order modes, although it will seen that this approach becomes exact either when the pump power is sufficiently large or sufficiently small. We therefore retain the Lagrange formulation, and substitute into (11) a Stokes field of the form:

$$\epsilon_s(\underline{r}) = A(z)\exp\left[-iQ(z)r^2/2\right] \tag{13}$$

The amplitude $A(z)$ and beam parameter $Q(z)$ are now chosen so that (11) is a minimum. Thus we carry out the transverse integrations, and apply the Euler-Lagrange equations for the variation of $Q^*(z)$ and $A^*(z)$:

$$\frac{3}{2}\,\frac{\frac{K_s^2}{n_s^2}\,\chi^{(R)}|\epsilon_{p0}|^2\,W_p^2(z)}{\left[4+iW_p^2(z)\,(Q-Q^*)\right]^2} = \frac{K_s\,Q'+Q^2}{\left[Q-Q^*\right]^2} \tag{14}$$

$$K_s\,\frac{d\,\mathrm{Ln}A}{dz} = \frac{|Q|^2+K_sQ'}{\left[Q-Q^*\right]} + \frac{3K_s^2}{4n_s^2}\,\frac{\chi^{(R)}\,W_{p0}^2|\epsilon_{p0}|^2\left[Q-Q^*\right]}{\left[4+iW_p^2(z)(Q-Q^*)\right]} \tag{15}$$

Equations (14) and (15) can be recast in terms of the normalised quantities as follows:

$$q^2 + \frac{dq}{d\zeta} + \frac{i\tilde{P}_p}{2\kappa^2}\left[1+\zeta^2-(\kappa\, \mathrm{Im}\, q)^{-1}\right]^{-2} = 0 \tag{16}$$

$$\frac{d\,\mathrm{Ln}\, A}{d\zeta} = \left[|q|^2 - \frac{dq}{d\zeta}\right]\left[q-q^*\right]^{-1} + \frac{\tilde{P}_p}{4\kappa}\left[1+\zeta^2-(\kappa\, \mathrm{Im}\, q)^{-1}\right]^{-1} \tag{17}$$

where:

$$\tilde{P}_p = \frac{3K_s^2}{2n_s^2} W_{p0}^2\, \mathrm{Im}(\chi^{(R)})|\epsilon_{p0}|^2 \tag{18}$$

$$q = \frac{K_p W_{p0}^2 Q}{2K_s} \tag{19}$$

$$\zeta = 2\left[\frac{z-f_p}{K_p W_{p0}^2}\right] \tag{20}$$

$$\kappa = K_s / K_p \tag{21}$$

$\tilde{P}_p$ is the 'normalised pump power' and ζ is the normalised longitudinal ordinate i.e. where possible, we have kept to the notation of [2] apart from the definition of $\chi^{(R)}$ which is that of [11]. Clearly if $\tilde{P}_p = 0$, (16) and (17) reduce to the equations of motion for the spot-size, radius of curvature, and (complex) amplitude of a free-space Gaussian beam. When $\tilde{P}_p \neq 0$ however, these equations can be used both to analyse the results of earlier authors in the domains of low and high pump power, *and* provide a more general description for the Stokes field for arbitrary $\tilde{P}_p$; these then are the respective goals of the sections which follow.

3. Solution to the equations of motion

3.1 Low pump power

We start by considering the limit of low pump power of the solutions to equations (16) and (17). We will first derive the general result for the Stokes amplitude and profile, and then show how this result can be applied to the design of a Raman gain cell.

If the normalised pump power $\tilde{P}_p$ is sufficiently small, the Stokes profile remains almost unchanged from its free space behaviour:

$$q^2 + \frac{dq}{d\zeta} = 0 \tag{22}$$

In terms of normalised quantities, the solution of (22) is:

$$q = \frac{\mu}{\mu(\zeta - \zeta_0) + i} \tag{23}$$

where: $$\zeta_0 = \frac{2F_s}{K_p W_{p0}^2} \tag{24}$$

and: $$\mu = \frac{K_p W_{p0}^2}{K_s W_{s0}^2} \tag{25}$$

Hence, ζ_0 is the distance of the Stokes focus from the pump focus in units of the pump confocal beam parameter, whilst μ is the ratio of pump to Stokes confocal beam parameter. This general case is depicted in figure 1 where the pump and Stokes beams have been enclosed by the gain medium. Of course, calculation of the Stokes field through equations (16) and (17) apply only to the field within the cell. Equally it is tacitly assumed that the finite transverse dimensions of the medium can be ignored.

With the free-space form for the Stokes profile, (17) can easily be solved to give the amplitude of the Stokes field at any point ζ in the gain medium:

$$A(\zeta) = A(\zeta_s)\left[\frac{1-i\mu(\zeta_s-\zeta_0)}{1-i\mu(\zeta-\zeta_0)}\right]\exp\left[\frac{\tilde{P}_p}{4\nu}\left[\tan^{-1}\frac{(\kappa+\frac{1}{\mu})\zeta-\mu\zeta_0}{\nu}-\tan^{-1}\frac{(\kappa+\frac{1}{\mu})\zeta_s-\mu\zeta_0}{\nu}\right]\right] \qquad (26)$$

where: $$\nu = \left[(\kappa+\mu)(\kappa+\frac{1}{\mu}) + \mu\kappa\zeta_0^2\right]^{1/2} \qquad (27)$$

and $A(\zeta_s)$ is the Stokes amplitude at the entrance to the medium. The total power in the Stokes beam can be evaluated from (13), (23) and (26):

$$P_s(\zeta) = P_s(\zeta_s)\exp\left[\frac{\tilde{P}_p}{2\nu}\left[\tan^{-1}\frac{(\kappa+\frac{1}{\mu})\zeta-\mu\zeta_0}{\nu}-\tan^{-1}\frac{(\kappa+\frac{1}{\mu})\zeta_s-\mu\zeta_0}{\nu}\right]\right] \qquad (28)$$

The justification for using the free-space profile (23) in deriving (28) is that our result for the Stokes power is then directly comparable with those of earlier workers. In fact Boyd et al [1] have obtained exactly the same result using an 'overlap integral' method. Whilst (28) is also related to the result obtained by Christov and Tomov [4]. Also, allowing for typographical errors, a similar result has been obtained by Trutna and Byer [3]. Examination of (16) reveals however, that to first order in $\tilde{P}_p$, the third term also contributes to the gain as described by (17). Thus we find that even for low pump powers, the effect of the pump power on the Stokes beam profile can be significant. However, this component can be shown to be identically zero for the particular initial Stokes profile satisfying: $\zeta_0 = 0$, and $\mu = 1$, which is just that the pump and Stokes beams share a confocal plane, and have equal confocal parameters. These are the conditions are chosen by Trutna and Byer to maximise their expression, based on (28), for the Stokes gain.

Strictly speaking, Trutna and Byer obtained a maximum gain through optimal choice of the confocal parameters belonging to both the pump and the Stokes field under the assumption that the beams share a confocal plane. They rightly concluded that the Stokes gain would be a maximum - in the limit of tight focussing for the pump - if the confocal parameters were equal. Thus in a cavity designed to give rise to a self-reproducing pump beam, the optimal choice of confocal parameters is also that which gives rise to a self-reproducing Stokes beam. We note in passing that for a cavity design other than that of Trutna and Byer wherein the pump beam is not tightly focussed, the condition $\mu = 1$ does not maximise the Stokes gain. In this case the characteristics and growth rate of the Stokes beam will be a result of the (competing) tendencies towards a beam that is self-reproducing, and one that has maximum gain. Within the variational framework of this chapter however, the condition above is a *necessary prerequisite* for the validity of (28). Therefore we will proceed assuming that these conditions are met by the design of the Raman amplifier, so that by virtue of our more general approach, we will then be in a position to determine the validity of the low gain approximation. In this case, the power gain for the Stokes beam is found to be:

$$P_s(\zeta) = P_s(\zeta_s) \exp\left[\frac{\tilde{P}_r}{2(1+\kappa)} \Theta(\zeta;\zeta_s) \right] \tag{29}$$

where:

$$\Theta(\zeta;\zeta_s) = \tan^{-1}\zeta - \tan^{-1}\zeta_s \tag{30}$$

If we now compare the magnitude of the discarded term in (16) with those used to define the free-space profile (23), then we find that the low gain approximation is consistent with the requirement:

$$\tilde{P}_p \ll 2(1+\kappa)^2(1+\zeta^2) \tag{31a}$$

which must therefore be regarded as a necessary condition for the validity of (29). In this form, the constraint above is rather unsatisfactory since it depends strongly on the length of the Raman gain medium. However, a more accurate constraint can be found from comparing the result (29), with an exact solution for the Stokes exponential gain which we will anticipate from the results of section 3.3. Thus by expanding the exponential gain in increasing powers of the normalised pump power, we find it is necessary that:

$$\tilde{P}_p \ll \left[32(1+\kappa)^5\right]^{1/2} \tag{31b}$$

whence in the low pump power domain, (29) gives an accurate measure of the Stokes exponential gain.

3.2 High pump power

As the pump power is increased, the Stokes profile will deviate from the free-space form given by (23). Hence the extent of the profile is governed by the competing effects of diffraction and gain-focussing determined respectively by the first and third terms of equation (16). When the pump power is sufficiently great, the effect of gain-focussing is to confine the Stokes spot-size to an area well within that of the 'guiding' pump, i.e. in the limit of high pump power we expect $w_p^2(z) \gg w_s^2(z)$. The Stokes spot-size can be defined using (13) and (19), in terms of the normalised variable q; whilst the pump spot-size can be defined using (7) and (20) in terms of the normalised co-ordinate ζ. Thus we may rewrite this condition as:

$$1 + \zeta^2 \gg \left[k \,\mathfrak{I}_m q\right]^{-1} \tag{32}$$

(If a TEM_{00} Stokes mode exists, then the imaginary part of q must always be negetive.) Using (32), (16) becomes:

$$q^2 + \frac{dq}{d\zeta} + \frac{i\hat{P}_P}{2k^2(1+\zeta^2)^2} = 0 \tag{33}$$

We note that the same result can be obtained by retaining only the zeroth and quadratic terms in the expansion of $|\epsilon_p|^2$ in powers of r^2 in (6). Hence this approach is just that of the parabolic-index profile approximation considered by Cotter et al [2]. However, in this paper we proceed to solve for q without the additional approximations made in that work.

Eqn. (33) is a Ricatti equation, and can be cast as a linear second order differential equation by making the usual change of variable:

$$q = \frac{1}{v}\frac{dv}{d\zeta} \tag{34}$$

also we define:

$$\gamma = \left[1 + \frac{i\tilde{P}_p}{2\kappa^2}\right]^{1/2} \tag{35}$$

whence:

$$\frac{d^2 v}{d\zeta^2} + \frac{(\gamma^2 - 1)}{(1+\zeta^2)^2}\, v = 0 \tag{36}$$

By substitution or otherwise, the solution of (36) can be shown to be:

$$v(\zeta) = v_0 \left[1+\zeta^2\right]^{1/2} \cos\left[\gamma\,\theta(\zeta;\zeta_s) + \phi\right] \tag{37}$$

where:

$$\theta(\zeta;\zeta_s) = \tan^{-1}\zeta - \tan^{-1}\zeta_s \tag{38}$$

and v_0, ϕ are (complex) arbitrary constants. From the definition (34), the complex parameter q can be recovered:

$$q(\zeta) = \frac{\left[\zeta - \gamma\tan(\gamma\theta + \phi)\right]}{\left[1+\zeta^2\right]} \tag{39}$$

where now ϕ can be interpreted in terms of the Stokes parameters at the entrance to the gain medium:

$$\phi = \tan^{-1}\left[\frac{\zeta_s - (1+\zeta_s^2)\,q(\zeta_s)}{\gamma}\right] \tag{40}$$

The amplitude of the Stokes field can now be obtained from (17), making use of (32):

$$\frac{d\,\mathrm{Ln}\,A}{d\zeta} = -q + \frac{\tilde{\rho}_p}{4\kappa(1+\zeta^2)} \tag{41}$$

Again, recalling the substitution in (34) and the result for $v(\zeta)$ in (37), the amplitude can be written down without further calculation:

$$A(\zeta) = A(\zeta_s)\left[\frac{1+\zeta_s^2}{1+\zeta^2}\right]^{1/2} \frac{\cos(\phi)}{\cos(\gamma\theta+\phi)} \exp\left[\frac{\tilde{\rho}_p\,\theta}{4\kappa}\right] \tag{42}$$

The condition (32) can now be stated using (39) as:

$$\mathrm{Im}\left\{\kappa\gamma\tan(\gamma\theta+\phi)\right\} >> 1 \tag{43}$$

Equations (39) and (42), in conjunction with (43), describe the behaviour of the Stokes field under the quadratic index profile approximation.

Consider now the initiation of the stimulated process from spontaneous scattering at $\zeta = \zeta_s$. If the initial field is a Gaussian beam with zero spot-size and zero radius of curvature, then we would have:

$$q(\zeta_s) = \infty - i\,\infty \tag{44}$$

After a short distance into the gain medium, the complex parameter q from (39) obeys:

$$q(\zeta_s) \simeq \frac{\zeta - i\gamma}{1+\zeta^2} \tag{45}$$

and the Stokes amplitude is:

$$A(\zeta) \simeq 2i\,A(\zeta_s)\left[\frac{1+\zeta_s^2}{1+\zeta^2}\right]^{1/2} \exp\left[\left(\frac{\tilde{\rho}_p}{4\kappa} + i\gamma\right)\theta\right] \tag{46}$$

The condition for the parabolic index profile approximation is now:

$$\kappa \, \mathrm{Im}\{\gamma\} \;\gg\; 1 \tag{47}$$

and in deriving (45) and (46), use has been made of the additional constraint:

$$\left| \exp\left[-2i\gamma\theta(\zeta;\zeta_s) \right] \right| \gg 1 \; ; \quad \zeta_s \leq \zeta \leq \zeta_e \tag{48}$$

This is a simplifying assumption designed to ensure that the cosine terms in (42) effectively collapse into the dominant exponential component. The value of ζ for which (48) becomes true depends on the magnitude of the gain: the higher the gain, the earlier will this constraint be satisfied and therefore will q approach the the particular form (45).

Defining the real and imaginary parts of Q in terms of the spot-size and radius of curvature (see for instance [12]):

$$Q \;=\; \frac{\kappa_s}{R_s(z)} \;-\; \frac{2i}{W_s^2(z)} \tag{49}$$

then we find that (45) implies that the Stokes beam has a radius of curvature:

$$R_s \;=\; \frac{\kappa_p W_{po}^2}{2} \left[\frac{1+\zeta^2}{\zeta + \mathrm{Im}\{\gamma\}} \right] \tag{50}$$

and spot-size given by:

$$W_s^2 \;=\; \frac{W_p^2(z)}{\kappa \, \mathrm{Re}\{\gamma\}} \tag{51}$$

Hence the Stokes field is a Gaussian beam with propagation characteristics similar to that of a free-space beam, but with a distorted phase front, and a spot-size that is everywhere narrower than its free-space equivalent.

Equations (45) and (46) describe the 'matched mode' behaviour of the Stokes field in that the complex parameter $q(\zeta)$ and amplitude $A(\zeta)$ have become independent of the initial parameter $q(\zeta_s)$. This is a generalisation of a concept first introduced in this context by Cotter et al [2]. The magnitude of the pump power, through the left hand side of (48), is seen to determine how quickly the initial Stokes profile tends towards the matched mode profile. In fact, if instead of (44), the initial parameter is made to satisfy the matched mode condition at $\zeta = \zeta_s$:

$$q(\zeta_s) = \frac{\zeta_s - i\gamma}{1 + \zeta_s^2} \tag{52}$$

then the $q(\zeta)$ remains unchanged from its matched mode value (45) throughout the medium.

These results can be compared with those of [2] by taking the high pump power limit for the complex parameter γ defined in (35). Under these conditions, the Stokes power is:

$$P_s(\zeta) \simeq 4 P_s(\zeta_s) \exp\left[\left(\tilde{P}_p - 2\sqrt{\tilde{P}_p} \right) \Theta(\zeta;\zeta_s) \Big/ (2k) \right] \tag{53}$$

where now (47) becomes:

$$\tilde{P}_p \gg 4 \tag{54}$$

Therefore, the results of [2] represent the high pump power limit of the matched mode solution. Note that by virtue of (54), the expression for the Stokes power in (53) is valid only when the net exponential gain is greater than zero. Thus the explanation based on this result which was advanced by Cotter for the behaviour of the Stokes beam at low pump power is spurious. Note also that (53) in conjunction with (54), describes a Stokes power similar to that obtained from the low pump power calculation of the previous section. The first term in the exponent is greater by a factor $(1 + \kappa)/\kappa$, whilst the additional second term represents a reduction in gain due to the increased diffraction of the Stokes field in the presence of gain-focussing.

3.3 Matched mode

Following the discussion in the previous section, we now seek an exact matched mode solution to the equations of motion (16) and (17) without making the parabolic index profile approximation. The result will then be an analytic description for the Stokes field that will be valid simultaneously under conditions of low pump power as for example in a multipass Raman gain cell, and conditions of high pump power likely to be encountered in a single pass Raman generator. In either case, the matched mode condition may be arrived at through one of two routes:

(a) An initially unmatched mode perturbed by the gain medium to a point where the spot-size and radius of curvature have converged upon that of the matched mode. From the previous section, we find this condition will generally be satisfied if:

$$\left| \exp\left[-2i\gamma\, \theta(\zeta;\zeta_s) \right] \right| \gg 1 \tag{48}$$

(b) An injected field which is a Gaussian beam with spot-size and radius chosen to satisfy the matched mode condition at $\zeta = \zeta_s$.

The (exact) matched mode solution to (16) may be derived from a substitution of the form:

$$q = \frac{\alpha - i\beta + e\zeta}{1+\zeta^2} \tag{55}$$

where α, β, and ϵ are real, and $\beta > 0$. Upon equating equal powers of ζ we obtain:

$$\epsilon = 1 \tag{56}$$

$$\alpha = \sqrt{\beta^2 - 1} \tag{57}$$

$$\beta \breve{P}_p = 4 (1 + \kappa\beta)^2 \sqrt{\beta^2 - 1} \tag{58}$$

whilst the amplitude now satisfies:

$$\frac{d \ln A}{d\zeta} = \frac{1}{[1+\zeta^2]} \left[\frac{\breve{P}_p \beta}{4(1+\kappa\beta)} + i\left(\beta + \frac{1}{\beta}\right)\right] - \frac{\zeta}{[1+\zeta^2]} \tag{59}$$

and so the Stokes power is easily found to be:

$$P_s(\zeta) = P_s(\zeta_s) \exp\left[G\left(\breve{P}_p, \kappa ; \theta\right)\right] \tag{60}$$

where $G(\breve{P}_p, k)$ is the matched mode exponential power gain:

$$G\left(\breve{P}_p, \kappa ; \theta\right) = \frac{\breve{P}_p \beta \theta}{2(1+\kappa\beta)} \tag{61}$$

and the matched mode complex parameter q is:

$$q = \frac{\sqrt{\beta^2 - 1} - i\beta + \zeta}{1 + \zeta^2} \tag{62}$$

and β is given by the solution of (58). That (60-62) encompass the results for high and low pump power of the previous sections can readily be seen from the limiting values for β. Thus if $\breve{P}_p$ is small, we can develop a series solution of (58) for β in increasing powers of $\breve{P}_p$ to obtain:

$$\beta = 1 + \frac{\breve{P}_p^{\,2}}{32(1+\kappa)^4} + \mathcal{O}\left(\breve{P}_p^{\,3}\right)$$

$$\text{and} \quad \Rightarrow \quad G = \Theta.\left[\frac{\breve{P}_p}{2(1+\kappa)} + \frac{\breve{P}_p^{\,3}}{64(1+\kappa)^6} + \mathcal{O}\left(\breve{P}_p^{\,4}\right)\right] \tag{63}$$

Alternatively, if $\breve{P}_p$ is large, then we can obtain a series solution for β in decreasing powers of $\breve{P}_p^{1/2}$ to obtain:

$$\beta = \frac{\breve{P}_p^{\,1/2}}{2\kappa} - \frac{1}{\kappa} + \mathcal{O}\left(\breve{P}_p^{\,-1/2}\right)$$

$$\text{and} \quad \Rightarrow \quad G = \Theta.\left[\frac{\breve{P}_p}{2\kappa} - \frac{\breve{P}_p^{\,1/2}}{\kappa} + \mathcal{O}\left(\breve{P}_p^{\,-1/2}\right)\right] \tag{64}$$

Note that comparison of the first and second terms in the expansions for the exponential gain, confirms the constraints for the validity of each of the results in the domains of low and high pump power in the previous sections (equations (31b) and (54) respectively).

We note in passing that the gain-focussed Stokes beam becomes ever more confined with increasing pump power and therefore can expect the parabolic index profile approximation discussed in section 3.2 to give increasingly accurate results. Thus the coupling between modes will eventually vanish and the high gain limit given by (60-62) with (64) will give the exact solution to (12). Further it is recalled that the result first obtained by Trutna and Byer [3] is effectively that of a first order perturbation theory (in the pump intensity) applied to a Stokes field expansion in free-space TEM_{nm} modes. Hence for sufficiently low pump powers, the limit given by (63) will also give an exact solution to (12).

For the general matched mode result, it may be of interest to know the spot-size and radius of curvature at any point ζ in the medium. Comparison of (45) with (55) reveals that the substitutions:

$$\mathrm{Re}\ \{\ \gamma\ \} \rightarrow \beta \tag{65}$$

$$\mathrm{Im}\ \{\ \gamma\ \} \rightarrow \alpha = \sqrt{\beta^2 - 1} \tag{66}$$

into (50) and (51) gives the general results:

$$R_s = \frac{K_p W_{po}^2}{2} \cdot \frac{1 + \zeta^2}{\zeta + \sqrt{\beta^2 - 1}} \tag{67}$$

(68)

where again β is given by the solution of (58). The radius and spot-size at the end of the gain medium can be found simply by substituting $\zeta = \zeta_e$ into (67) and (68) respectively. It is clear from these results that the radius of curvature and the spot-size are smaller than that of the equivalent free-space mode which has $\mu = 1$ and a shares a focal plane with the pump beam.

4. Discussion

We now compare the predictions of (60-62) with both the approximate results of sections 3.1 and 3.2, and the results of [5]. In the following, we will assume that the pump focussing conditions are such that $\theta(\zeta;\zeta_m) \simeq \pi$, and therefore that the Stokes exponential power gain is given by:

$$G(\breve{P}_p, \kappa) = \frac{\breve{P}_p \, \beta \, \pi}{2\,(1+\kappa\beta)} \tag{69}$$

The equation (58) has been solved for β numerically, and a plot of β versus $\breve{P}_p$ for various values of κ is given in figure 2. These results can be used to find the matched mode exponential gain $G(\breve{P}_p,\kappa)$ in (61) and the spot-size and radius of curvature in (67) and (68). In figure 3 we compare the matched mode gain with the gain predicted by (63) and (64). As expected, it is seen that the limiting cases are satisfactorily modelled as $\breve{P}_p \to 0$ and $\breve{P}_p \to \infty$ respectively. For the chosen value of $\kappa = 1$, we observe that the predictions of the low and high gain approximations are equal at a pump power $\breve{P}_p = 16$ (the high pump power solution thereafter being closer than the solution for low pump power to that of the matched mode). In this respect then, this is the point at which the conventional models are least satisfactory - there being about 15% deviation from the matched mode gain.

It is also of interest to compare these results with those obtained by Perry et al [5] (see figure 1 of that work). First it is necessary to make explicit the connection between the symbols used in their work, and those adopted in this paper. Table 1 provides a summary of the pertinent relationships:

Table 1

Description	Perry et al	This paper
Dimensionless parameter	ρ	$\kappa/(1+\kappa)$
Gain coefficient	G_ρ	$\hat{P}_\rho/4$
Real part of eigenvalue	$Re\{\lambda\}$	$\breve{P}_\rho\,\beta/(4\,(1+\kappa\beta))$
Normalised gain	$Re\{\lambda\}/G_\rho$	$\beta/(1+\kappa\beta)$

In figure 4 we have used these relationships to compare the matched mode gain with the gains predicted in [5] (at $\kappa = 1$) for the first and second (rotationally symmetric) eigensolution. Clearly the matched mode gain is consistently close to the gain of the first eigensolution and the excellent correspondence between these results lends support to our model. Encouraged by this comparison, we present in figure 5 the matched mode gain for various values of κ found by applying the solution of (58) to (61).

It is possible to further test the accuracy of our supposition that the lowest order mode is essentially a Gaussian beam by comparing the matched mode profile with that predicted by the numerical results of [5]. With reference to figure 6, we have used the normalised coordinate $r/w_s(z) = r\sqrt{k\beta}/w_p(z)$, and find that once again, at least for the values $\hat{P}_p = 40$ and $k = 1$, there is very good agreement between the results.

5. Summary

In this paper we have presented an analytic model for the evolution of a Stokes field in a Raman active medium excited by a focussed pump beam. We have shown that our results are valid throughout a wide range of values for the pump power, and that in the limits of high and low pump power, they reproduce the results of earlier workers. We have therefore been able to identify constraints which in this context define the domains of high and low pump power. Excellent agreement has been obtained in comparison with an earlier numerical treatment.

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Legends for figures

Figure 1: Configuration of pump and Stokes beam in a gain medium

Figure 2: Dimensionless parameter β versus normalised pump power $\widetilde{P}_p$.

Figure 3: Stokes exponential gain as predicted by matched-mode, low-gain, and high-gain theory.

Figure 4: Stokes exponential gain as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.

Figure 5: Stokes exponential gain as predicted by matched-mode theory for k = 0.1, 0.5 and 1.0.

Figure 6: Stokes profile as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.

FIGURE 1

Configuration of pump and Stokes beam in a gain medium.

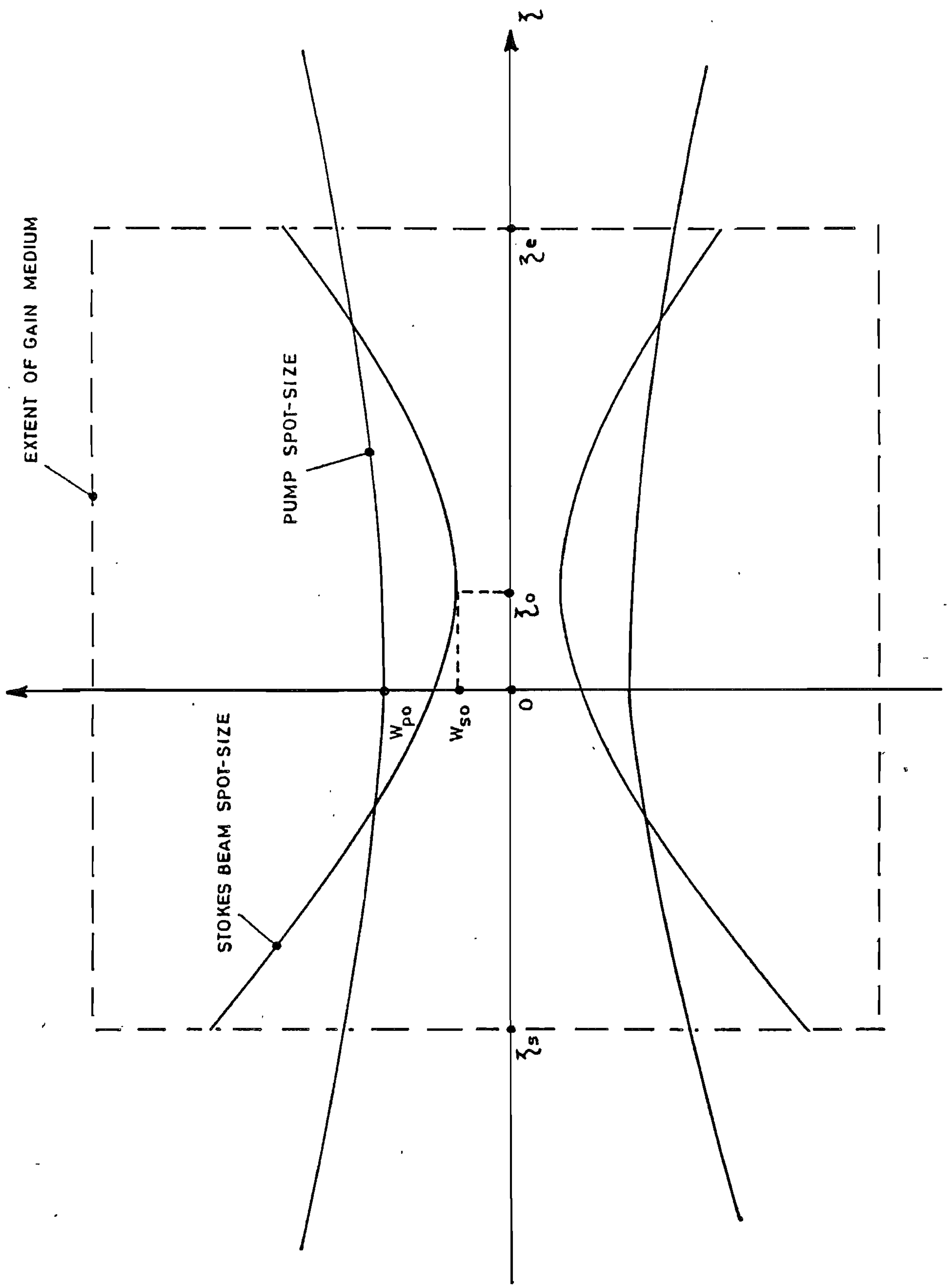


FIGURE 2

Dimensionless parameter β versus normalised pump power for various κ.

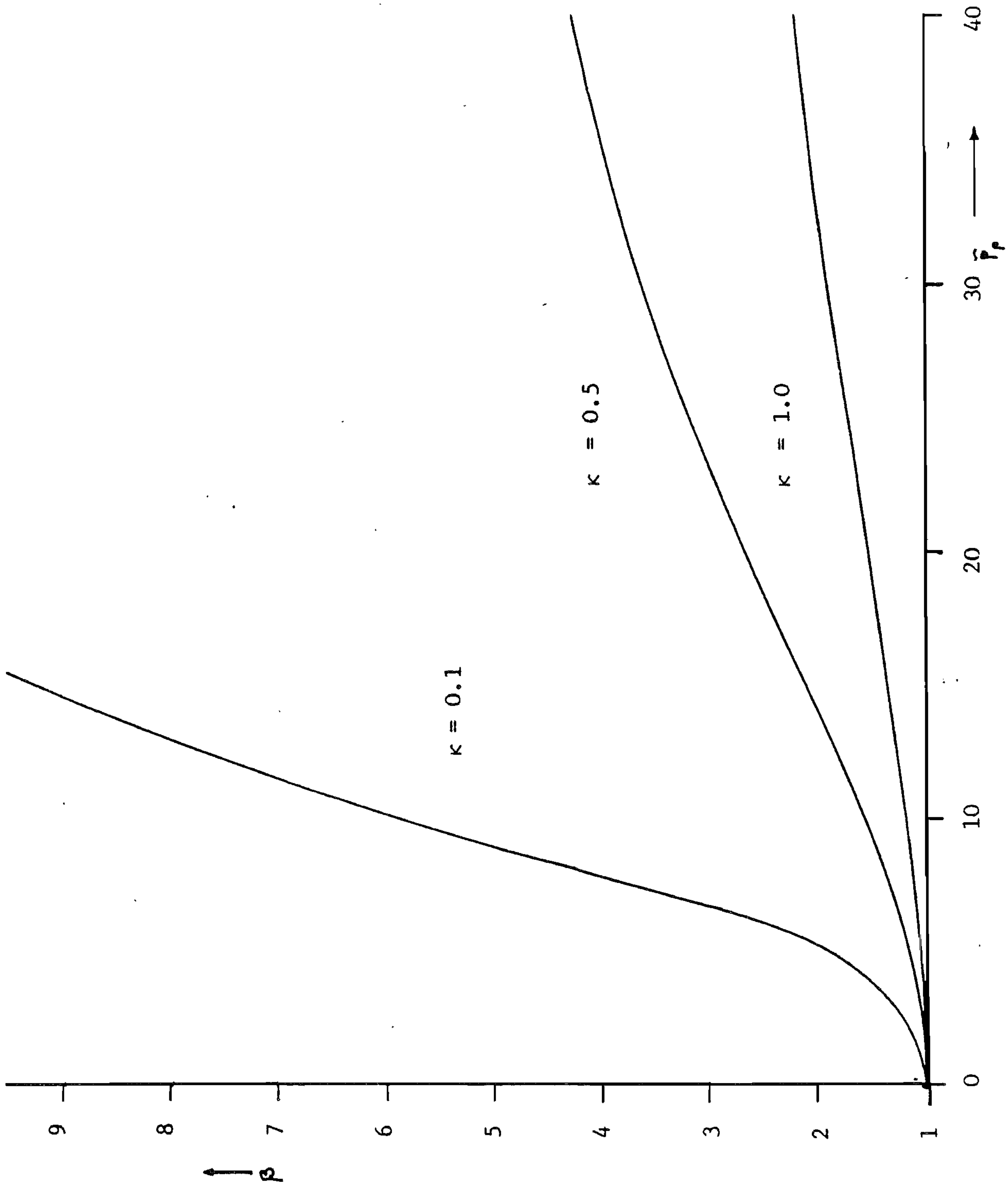


FIGURE 3

tokes exponential gain as predicted by matched-mode, low gain, and igh gain theory.

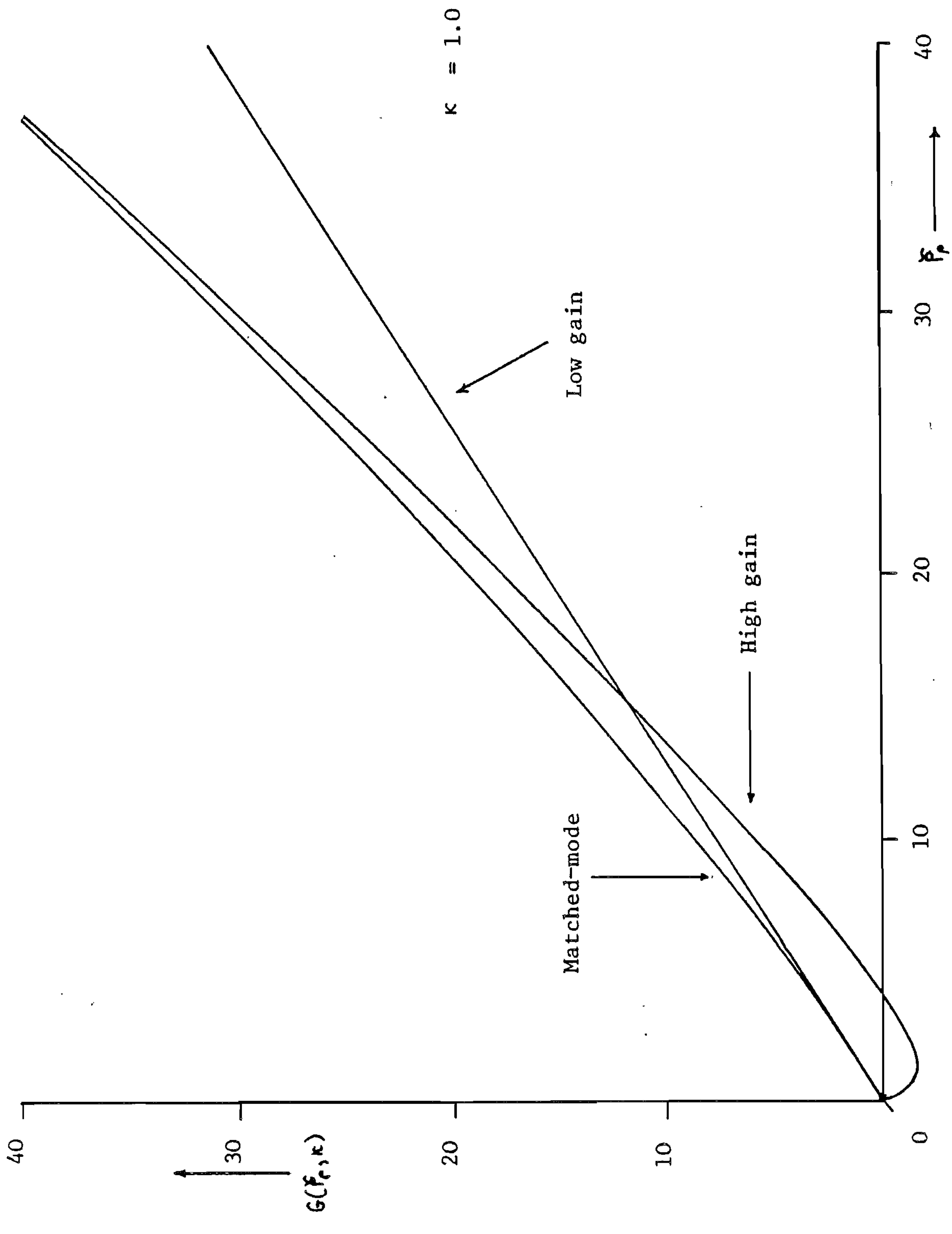


FIGURE 4

Stokes exponential gain as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.

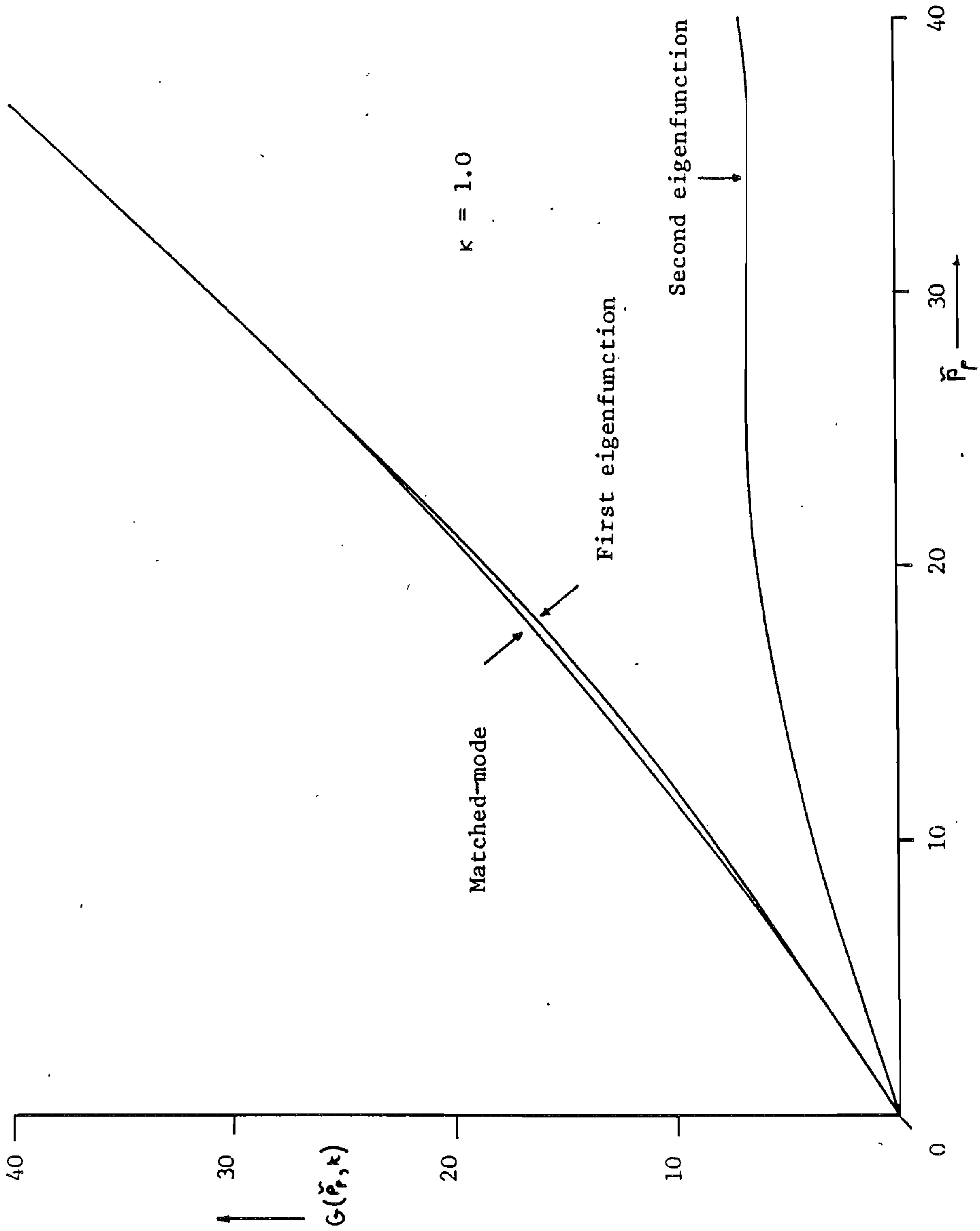


FIGURE 5

Stokes exponential gain as predicted by the matched-mode theory versus normalised pump power for various κ.

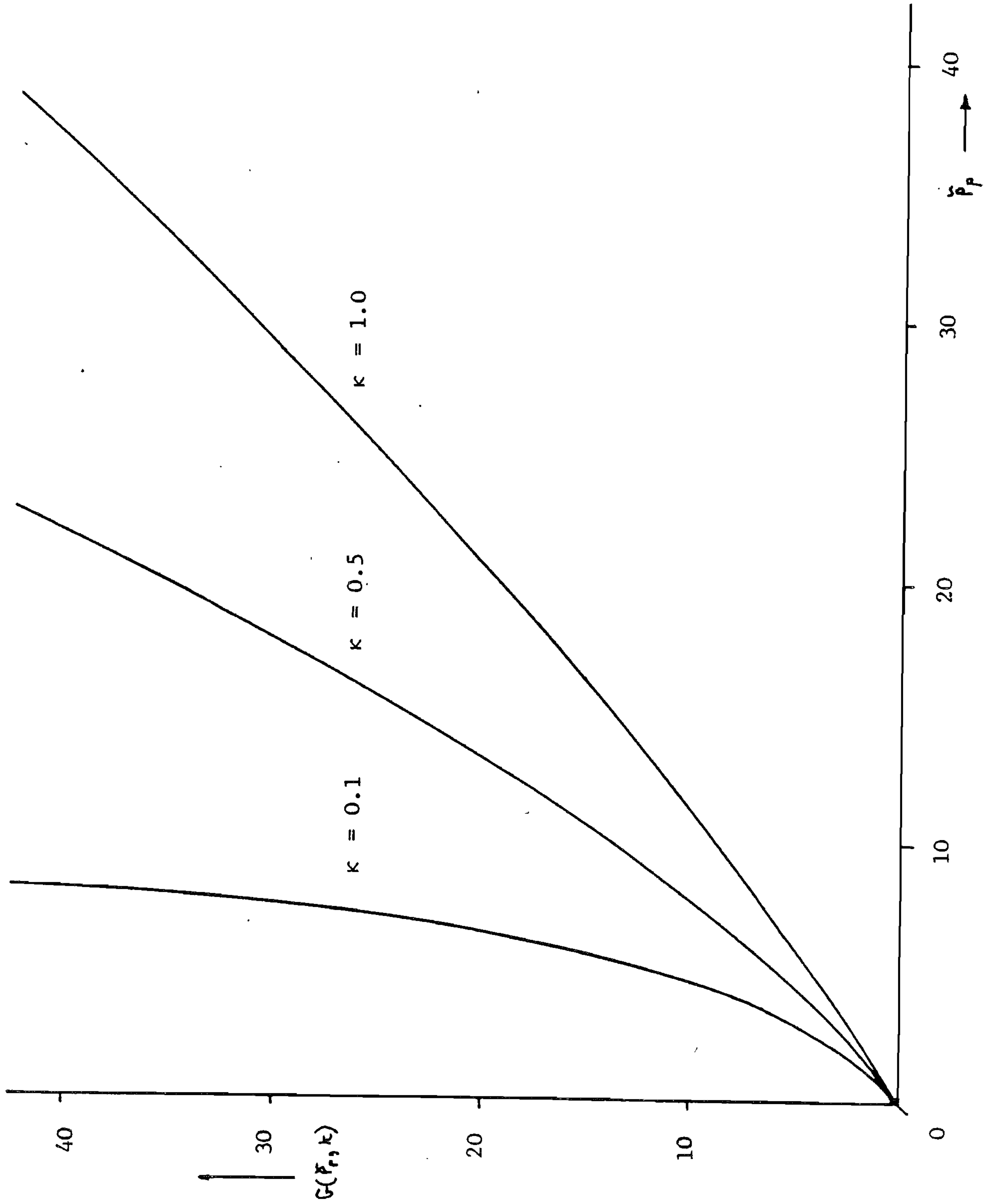
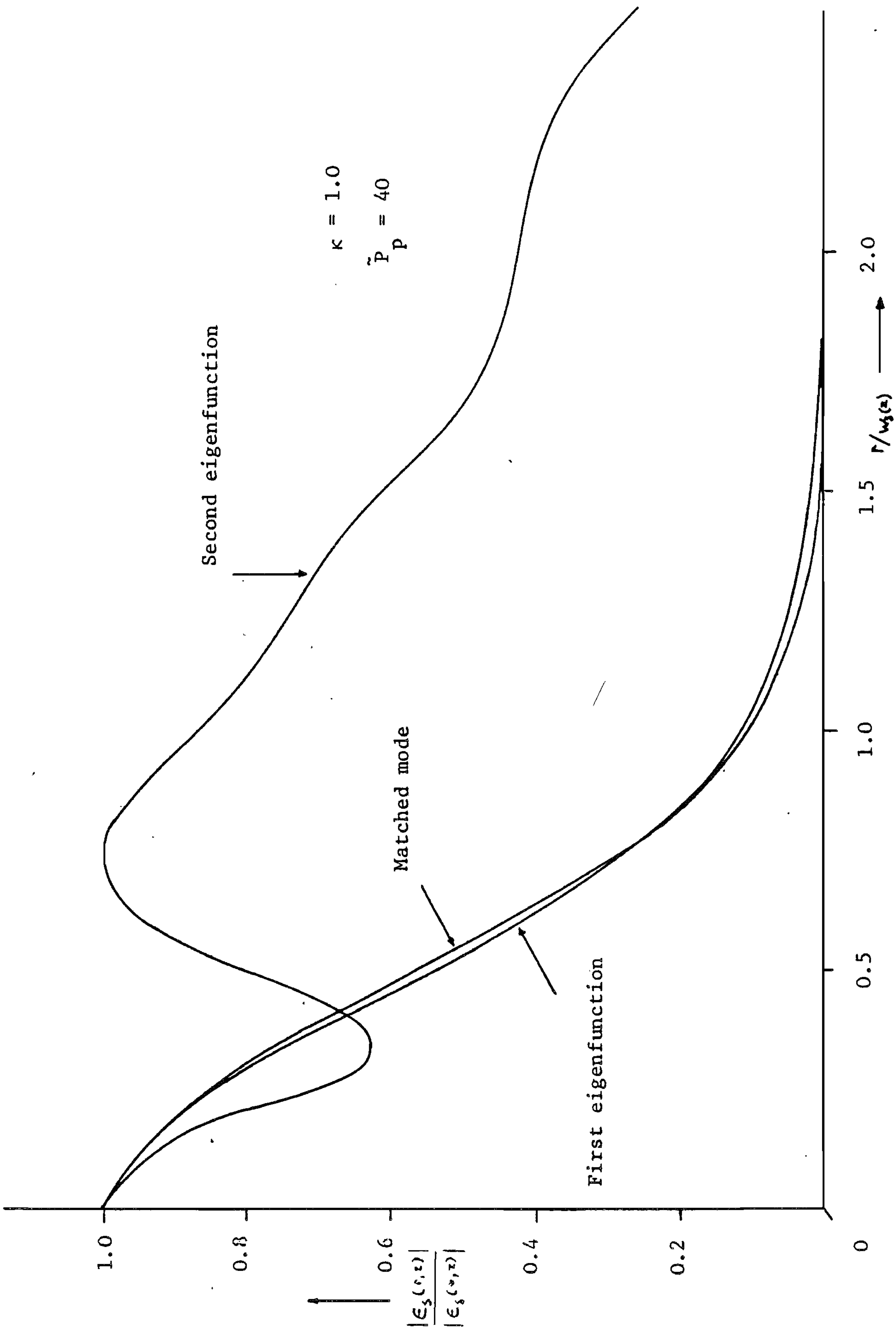


FIGURE 6

Stokes profile as predicted by matched-mode theory and that of first and second eigenfunctions of Perry et al.



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