1 January 1995

OPTICS COMMUNICATIONS

ELSEVIER

Optics Communications 113 (1995) 550–558

Full length article

Anomalous photorefractive response in two-beam coupling with focussed beams in photorefractive crystals and waveguides

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Received 14 July 1994

Abstract

We report on anomalous two-beam coupling results observed in photorefractive $BaTiO_3$ crystals and ion-implanted waveguides when focussed beams have been used. The two-beam coupling gain, investigated as a function of the input beam ratio, showed an enhanced magnitude, when compared with the standard theory, and exhibited a strong beam ratio dependence. In order to explain the significant difference we observed between theory and experiment, we discuss several models, based on intensity-dependent processes, and show that only a modification with the enhanced photoinduced space-charge field being a nonlinear function of modulation, has given an agreement with the experimentally determined results.

1. Introduction

Over the past two decades many interesting phenomena have been observed using photorefractive materials. Effects like optical phase conjugation (generated in either externally pumped or self-pumped configurations) and various multiple wave mixing arrangements have been extensively investigated, both experimentally and theoretically. However, the prodigious advances in knowledge of photorefractive optics is currently not matched by development of practical devices, which requires media exhibiting not only large nonlinearities, but also fast response times. Photorefractive materials like $BaTiO_3$, display strong nonlinear effects, but, unfortunately, they also have slow response time. There have been several schemes suggested to decrease their response time such as the manipulation of ion impurity concentration [1], use of elevated temperatures [2], and increased input beam intensity. This third method can be achieved by simply increasing the laser output or, equivalently, by focussing the input beams. Better still however, is to adopt a waveguide structure in the photorefractive material. Such waveguides can be fabricated by implanting light ions (He^+ and H^+) into a polished crystal [3]. These high energy ions, gradually slowed down by electronic and nuclear interactions, produce a distinct layer of lower refractive index buried 5–15 μm below the crystal's surface. In this way, light can be confined in a planar waveguide, created between the polished top face of the crystal and the reduced index barrier.

Photorefractive waveguides should not only respond much faster to a given incident power of the incident light, but also preserve the crystal's intrinsic electro-optic properties, and ideally exhibit low losses. The properties of the waveguides were investigated using three main techniques: two-beam coupling, self-pumped phase conjugation, and mutually pumped

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SSDI 0030-4018(94)00573-7

phase conjugation [3–5]. The phase conjugate reflectivity, and two-beam coupling gain were measured, together with their response times. It was additionally expected that ion implantation would affect the photorefractive properties of a given a material. The results achieved [3–5], indicated that the response time decreased by two orders of magnitude, and that the direction of the two-beam coupling gain was reversed when compared with bulk crystals. Since these first interesting results, work has been undertaken to establish a more systematic characterization of the waveguide properties. Using the two-beam coupling technique, with beams focussed in order to launch them into a waveguide, we have studied the behaviour of the gain experienced by one of the beams. The data obtained, however, showed an unexpected difference from the standard theory; to find the source of this discrepancy, the same two-beam coupling experiment with focussed beams was repeated in bulk crystals, and again similar anomalous results were obtained. Hence, it could be concluded that the modified photorefractive properties were not alone responsible for the anomalous beam coupling behaviour. Our aim, therefore, was to produce a theoretical model which, accounting for a particular geometry of two-beam coupling in a waveguide, would provide a good description of this process and hence help to determine the waveguide parameters.

The following sections will be devoted to a more detailed study of the two-beam coupling gain. The anomalous features of this phenomenon will be discussed, and the results obtained from theoretical modelling will be compared with the experimental results.

2. Two-beam coupling as an experimental technique

Two-beam coupling describes the phenomenon in which two beams overlap in a photorefractive crystal, and one experiences gain at the expense of the other, which gets depleted. Since this effect has been widely researched, it seemed ideally suited as a method to quantify the photorefractive material parameters.

The characterisation of the effective two-beam coupling gain involves measurements of its dependence on the input beam power ratio. The effective gain G, i.e. the probe beam amplification, can be defined as

$$G = \frac{I_s \text{ with } I_p \text{ present}}{I_s \text{ without } I_p \text{ present}}, \tag{1}$$

where I_s is the signal beam, and I_p is the pump beam.

The expression for the gain can be calculated analytically from the wave equations describing the interaction between two plane waves inside a photorefractive medium [6]. If negligible absorption is assumed, the gain G is expressed by

$$G = \frac{(1+r)e^{\Gamma L}}{1+r e^{\Gamma L}}, \tag{2}$$

where Γ is the coupling coefficient, $r = I_s/I_p$ is the input beam power ratio, and L is the effective interaction length.

It can be shown that for $r \ll 1$, the effective gain yields the value of the coupling coefficient: $\Gamma = \ln G/L$. Hence, a measurement of the effective gain provides the actual magnitude of the coupling coefficient. This coupling coefficient can also be determined theoretically from the geometry of the interacting waves and the material parameters [6]:

$$\Gamma = \frac{2\pi}{\lambda \cos\theta} r_{\mathrm{eff}}\, n_0^3\, |E_w|, \tag{3}$$

where 2θ is the internal crossing angle between the incident beams, n_0 is the unperturbed refractive index of the material, and E_w is the modulation-independent part of the fundamental component of the induced, steady state space-charge field E_{s1} [7]

$$E_{s1} = E_w m, \tag{4}$$

where modulation $m = 2\, \boldsymbol{E}_s \cdot \boldsymbol{E}_p^*/(I_s + I_p)$ and $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ are slowly varying amplitudes of pump and probe beam, respectively. In the absence of externally applied fields E_w is equal to

$$E_w = \frac{E_q E_D}{E_q + E_D}, \tag{5}$$

where r_{eff} denotes the effective electro-optic coefficient [8], $E_D = k_B T\, K/e$ is the diffusion field, and $E_q = eN_{D_{eq}}^{+}/\epsilon\epsilon_0 K$ is the trap-limited saturation field strength, k_B is Boltzmann's constant, T is the temperature in kelvin, K is the magnitude of the grating vector $\boldsymbol{K}$, e is the magnitude of the electronic charge, $N_{D_{eq}}^{+}$ is the concentration of ionised donor-like trapping centres in quasi-equilibrium under dark conditions, ϵ is

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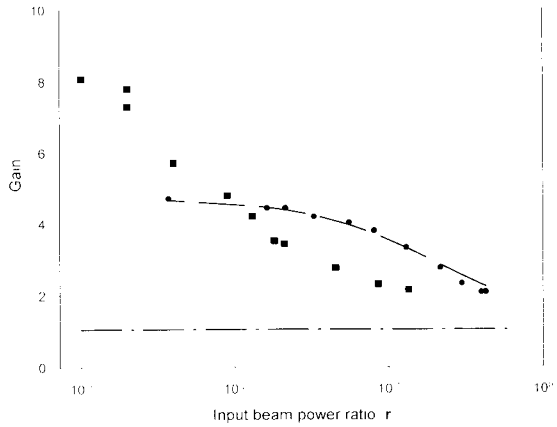

Fig. 1. Two-beam coupling gain as a function of the input beam power ratio: – for the case of collimated beams in a bulk crystal, $\beta = 0°$, $\theta = 4°$, $\Gamma L = 1.6$, •: experimental data, dashed line: standard two-beam coupling theory curve. – for the case of focussed beams launched into a waveguide, $\beta = 0°$, $\theta = 24°$, $\Gamma L = 2.13$, ▪: experimental data, dashed–dot line: standard two-beam coupling theory curve.

the static dielectric constant of the crystal, and ϵ_0 is the free space electric permettivity.

Hence, the exact experimentally determined value of the coupling coefficient Γ provides invaluable information about the parameters of the photorefractive material; in our case an important indication of the performance of the photorefractive waveguide.

The simple two-beam coupling experiment, performed in a crystal with collimated Gaussian beams, usually gives a good agreement between theory and experiment. As an example, Fig. 1 includes the recorded experimental data (circles), and the theoretical prediction (dashed line), for two-beam coupling gain in a single 5.8 × 5 × 5 mm^3 $BaTiO_3$ crystal. The crossing angle of the two beams was about 4° and the grating wave vector was parallel to the c axis. The theoretical gain curve was obtained using Eq. (2) with Γ as an adjustable fitting parameter. The value of Γ obtained in this way agreed within 2% with the value calculated from Eq. (3), using known parameters of the particular $BaTiO_3$ crystal. The presence of strong beam fanning in this classical two-beam coupling experiment can sometimes complicate interpretation of the beam coupling [9], so particular care is required to suppress the fanning gratings to measure the exact value of the two-beam coupling gain alone.

3. Anomalous two-beam coupling results

The experimental studies of two-beam coupling were performed initially in photorefractive waveguides and then, subsequently for comparison, in bulk crystals. An Ar^+ laser operating at λ=488 nm was used to launch extraordinary polarised light into a 15 μm deep and 5 mm long H^+ implanted waveguide, the beams being tightly focussed by two × 4 microscope objectives. The internal crossing angle ($\theta = 24°$) and the angle between the grating $\boldsymbol{K}$ vector and the c axis ($\beta = 0°$) were kept fixed. Outputs from the waveguide region were imaged, via subsequent microscope objectives, onto calibrated optical power meters. Owing to the focussed beam geometry no significant beam fanning was observed.

The results obtained (marked as squares in Fig. 1), showed an apparent strong dependence on the input beam ratio. This dependence was not predicted by the standard theory (marked as dashed–dot line), Additionally, the gain observed was approximately eight times larger than expected. The same, or similar, results were obtained in all the photorefractive waveguides studied. To investigate the origin of this anomaly, the same two-beam coupling experiment, using focussed beams, was performed in bulk crystals. The gain showed similar large discrepancies compared with theory indicating that the anomalous two-beam coupling effect was not due to particular properties of the waveguides, but was the result of either the high optical power density used or the specific geometry adopted for the interaction.

The main difference between previous experiments [10,11] and the ones described here is the small size (down to approximately 10 μm diameter) of both interacting beams. Using focussed beams for two beam coupling, enables a dramatic improvement in the response time, but is usually accompanied by a small value of gain, due to the consequently limited interaction region. The unexpected, significant increase of gain observed in our anomalous two-beam coupling opens up the possibility of obtaining simultaneously both efficient, and fast coupling between two laser beams, and is therefore worth a thorough investigation.

A set of experiments was performed to check the dependence of the two-beam coupling gain on the input beam ratio for: different crossing angles (large $\theta = 24°$, and small $\theta = 4°$), different inclination of the crystal versus incident beams ($\beta = 0°$: grating vector $\boldsymbol{K}$ parallel to the c axis of the crystal, and $\beta = 15°$), and also for different sizes of focal spots of incident beams. The precise geometry of beams inside the crystal was carefully monitored via a camera mounted above the crystal.

For all cases two important coupling parameters were required: the value of saturated gain, necessary to obtain the value of coupling coefficient Γ, and the profile of the gain dependence versus the input power ratio. At first, we checked the case of collimated (2.5 mm in diameter) and slightly focussed (1 mm in diameter) beams. The magnitude of the coupling coefficient deduced from the measurements agreed quite well (within 5%) with the value calculated from Eq. (3). This agreement was obtained for all experimental arrangements we considered. For a small crossing angle, as for example shown in Fig. 1, the standard theory fitted the experimental data quite well. However, for the large crossing angle ($\theta = 24°$), we found that the fit to the experimental data was not as good. This discrepancy could, however, be explained and corrected by modifying the two-beam coupling theory to account for the Gaussian profiles of the interacting beams [12].

After this preliminary investigations, we concentrated on the case of strongly focussed beams, crossing at the large angle $\theta = 24°$. For this case, it was observed that the gain didn't saturate even for very small input power ratios ($r = 10^{-6}$), and that its maximum value was much higher that the one calculated from the theory. This deviation from the theoretical value increased with tighter focusing, reaching a factor of approximately ten for the 100 μm diameter focussed beams. At the same time, the discrepancy between the experimental and the standard theoretical dependence of gain on the input power ratio increased by up to a factor of 5×10^3, in the most extreme case. In fact, this factor was much higher that the one obtained in the waveguide measurements. This anomalous behaviour was common to several sets of data, and was obtained in all repeated experiments. It could be shown that this anomalous behaviour could not be explained, as in the case of collimated beams, just by adopting Gaussian profiles. We therefore attempted to model this unusual photorefractive interaction, starting with the analysis of various intensity and spatially dependent phenomena associated with two-beam coupling.

4. The effect of intensity dependent processes in two-beam coupling

It has become evident, from the experimental results obtained, that the standard coupled-wave theory, had to be modified. From the observed dependence of the gain on the input power ratio, the possibility that the product ΓL, was a function of the illumination intensity must be considered, despite the fact that the theory predicts (Eq. (3)) that the coupling coefficient should be independent of the input power. In fact, even the earliest measurements of Γ showed that it does indeed change as the input power ratio is varied [13], but this intensity dependence has usually been neglected.

More recently the intensity dependence of various photorefractive properties has been extensively studied. It has been found that effects like absorption [14] or parameters including space-charge field, photoconductivity [15] or effective trap density [16] could all be nonlinear functions of the incident intensity. We have considered physical mechanisms giving rise to these phenomena, and their possible contribution to the effect we observe.

Most of the intensity dependent effects can be accounted for by the presence of secondary traps [16]. Tayebati and Mahgerefteh [17] presented a detailed model of the photorefractive effect which consisted of a two-level system model, and included optical and thermal excitation from both trap levels [18]. According to this model, photogenerated charges acquired by shallow traps form a charge grating, which strongly influences the total space-charge field. Therefore, the possibility exists that these secondary trap centres in our crystals lead to the anomalous two-beam coupling. Experimentally, the influence of the secondary traps manifests itself most clearly in the asymmetry of beam coupling with respect to the orientation of the c axis, which originates from the existence of additional absorption gratings. The electro-optic gratings also get modified and this is expressed as

$$E_{s1m} = \eta(I_0) E_{s1} , \tag{6}$$

where E_{s1} is given by Eq. (4), I_0 is the total incident intensity, and $\eta(I_0)$ is a nonlinear function of the total intensity [18].

It can be shown that the presence of shallow traps reduces the space-charge field by a factor η which varies between $1/2 \leq \eta \leq 1$. Hence, by definition, the secondary levels cannot explain the significant increase of coupling observed for the tightly focussed geometry. Moreover, using a simple model it can also be proved that the formula for $\eta(I_0)$ [18] predicts a different kind of gain intensity dependence than the one observed in our experiment.

Considering the absorption gratings, it can be calculated that their contribution to the total coupling would be small, i.e. too small to explain a significant increase in gain. In parallel with this theoretical analysis, a simple experiment on the light-induced absorption proved that, in fact, the effect observed turned out to be small, i.e. the change of absorption with input power ratio was smaller than 10%, which only confirmed the conclusions reached from the theoretical model, that the change in the two-beam coupling was not caused by secondary trap centres.

The intensity-dependent behaviour in $BaTiO_3$ has also been observed in two-beam coupling experiments with high-intensity (few MW/cm^2) pulsed illumination [19], arising from the photocarrier saturation and change in the relative contributions of hole and electron conductivity. In our case, however, the power density did not exceed 20 kW/cm^2, so our experimental regime was well below the one required for invoking electron-hole competition.

As can be seen from the expression (2), the gain is determined by the product of the coupling coefficient, Γ, and the interaction length, L. Hence, we also investigated the possibility of a change of the overlap region due to nonlinear effects like self-focusing and self-trapping. It has been reported that self-trapping may be observed in photorefractive crystals only under certain experimental conditions [20], which are, again, quite different from ours. In order to justify the significant increase in gain, the increase in the interaction length would have to be quite substantial. Such effects were not observed in routine monitoring using a camera mounted above the crystal. However, in order to rule out the possibility of any change in length of the interaction region with intensity, an experiment was performed, where the output beam diameter was measured on a beam profiler for different input beam intensities. No significant variation of the spot size was detected. Hence, we were able to conclude that it was the change in the coupling coefficient, which was responsible for the anomalous gain, rather than the change in the effective interaction length.

In summary, our analysis indicated that no intensity dependent processes were responsible for the anomalous two-beam coupling behaviour observed.

5. Photorefractive response in a finite interaction region

5.1. Interaction geometry

In order to understand better the energy transfer occurring in the particular arrangement of our experiment, we re-examined the assumptions and approximations used to describe the process of grating formation and the coupling between the beams. We have, therefore, concentrated on establishing the importance of our particular experimental geometry, including the consequences of using small diameters of the beams (from 2.5 mm to 10 μm), and therefore on having an interaction region occupying only a small fraction of a crystal volume. Simple calculations showed that in spite of the fact that the overlap area in our experiment was limited, it was large enough to fulfil the conditions for the slow exchange of energy between the beams. The interaction zone was 2–3 orders of magnitude larger than the grating period, and the angular diffraction spread was small compared to the angle of propagation θ. Hence, even tightly focussed beams could be regarded as collimated within the interaction region, which contained at least 250 fringes. In the next step, we have studied the shape of the intensity interference pattern formed by two focussed beams, which could be characterised by Gaussian profiles. As was shown, the total optical intensity pattern formed by two Gaussian beams also acquires the shape of a Gaussian distribution [21,22]. This relation is, in fact, valid for symmetrical waves with any spatial finite extent. Further analysis of the steady state energy coupling for Gaussian optical beams showed that photorefractive gain does not vary exponentially with distance through the crystal, as in the case of plane waves, but the coupling coefficient Γ is exactly the same. Two-beam coupling theory for Gaussian beams, which also has been reported recently [23], yields, as expected, that the gain has to be lower for two Gaussian beams than for two plane waves due to a shorter interaction region, but beyond that, it does not predict any anomalous effects. Furthermore, the excessive gain we observe is not accounted for. Indeed, as we have already mentioned, the attempts to explain our experimental data by using the standard two–beam coupling model with Gaussian profiles of interacting waves failed to give a good agreement [12].

5.2. *Perturbation analysis and an empirical correction function*

Using our conclusions about the shape of the interference profile and the modulation pattern created inside our photorefractive material, we propose the following explanation of the anomalous effects. Strong coupling occurring between focussed beams over a short interaction region must originate from the modified strength of space charge field. Comparison between the experimental data and the standard plane wave or Gaussian theory, which predicts low gain, indicates the presence of some enhancement mechanism that gives rise to $E_w > E_q$ at small values of input beam ratios r. This mechanism is only pronounced at high input intensities, a condition obtained via focussed beams. For large values of beam ratios, the gain decreases and is determined by the field E_q, as expected, i.e. approaching the low–gain standard theory fit. Such behaviour is in fact similar to the effects observed in non-stationary recording [24]. Non-stationary conditions could be the source of nonlinear modulation of the space charge field, and therefore the necessity of incorporating second order perturbation correction had to be checked.

Standard charge transport equations are described theoretically by a set of nonlinear differential equations, which express the photorefractive space-charge field E_{sc} as a function of the light intensity pattern $I(\boldsymbol{r})$. In general, however, an exact analytical solution of these equations cannot be found. The most common approach, therefore, is to linearise the equations, so the incident intensity then takes the form of a one-dimensional sinusoidal pattern created by the interference of two plane waves

$$I(\boldsymbol{x}) = I_0\{1 + \mathrm{Re}[m\exp(\mathrm{i}\boldsymbol{K}\cdot\boldsymbol{x})]\}. \tag{7}$$

Given this form of $I(x)$, the standard theory predicts that the fundamental component of the space charge field is linear with the modulation depth m (Eq. (4)). The solution for $E_{sc}(x)$ is also strictly valid only for small m. However, the accuracy of this result depends strongly on the conditions under which the internal space-charge field is created. Any evidence that the coupling coefficient changes its magnitude with the modulation index m implies, that the space-charge field must vary nonlinearly with the modulation index. Experimentally it has been shown that such a nonlinear photorefractive response can be observed in many photorefractive materials for various external conditions; externally applied AC-fields or the moving grating technique are the most common causes of the finite higher-order components of the space-charge field [25–27]. Additionally, it has been recently reported that the perturbation correction had also been used to explain some data obtained in $BaTiO_3$ with no external fields applied [28,29].

Several perturbative approaches, with different empirical forms of the nonlinear response function, $f(m)$, have been presented to obtain a higher-order analytical expression for the steady-state space-charge field as a function of modulation depth m:

$$E_{s1} = f(m)E_{w1} \tag{8}$$

Using the form of $f(m)$ as in Ref. [25]:

$$f(m) = \frac{1}{a}[1 - \exp(-am)] \tag{9}$$

the coupled differential equations describing the propagation of two incident beams inside the photorefractive material become

$$\frac{\mathrm{d}I_p}{\mathrm{d}x} = -\Gamma\frac{f(m)}{m}\frac{I_pI_s}{I_p+I_s},$$
$$\frac{\mathrm{d}I_s}{\mathrm{d}x} = \Gamma\frac{f(m)}{m}\frac{I_pI_s}{I_p+I_s} \tag{10}$$

so the new coupling coefficient may be treated as the product of the old Γ and the ratio of the new perturbation correction function to the modulation depth. The main advantage of defining $f(m)$ as in Eq. (9) is that after introducing the simplifying assumptions of no absorption and no depletion of the pump beam, the solution for gain, G, in the signal beam can be easily derived analytically [25] giving

$$G = \frac{1}{4a^2r}\left[\ln\left(1 + [\exp(2a\sqrt{r}) - 1]\exp\frac{\Gamma L}{2}\right)\right]^2. \tag{11}$$

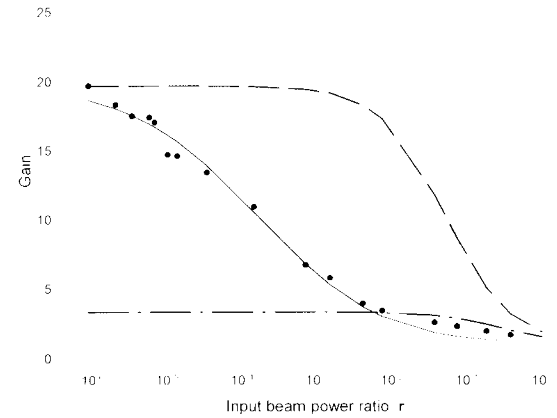

Fig. 2. Two-beam coupling gain as a function of the input power ratio for the case of beams focussed to $2w_0 = 400\ \mu m$ in diameter in a bulk crystal $\beta = 0°$, $\theta = 24°$, $\Gamma L = 3$, $a = 11$ •: experimental data, solid line: theoretical fit with the nonlinear response empirical correction function, dashed–dot line: standard theory curve for the value of $\Gamma L = 1.2$, (value expected from the standard theory) dashed line: standard theory curve for $\Gamma L = 3$.

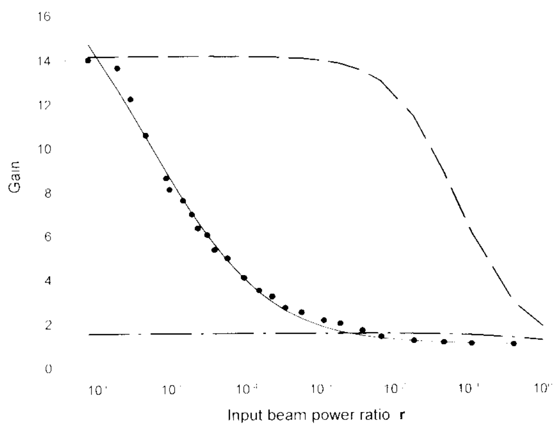

Fig. 3. Two-beam coupling gain as a function of the input power ratio for the case of beams focussed to $2w_0 = 100\ \mu m$ in diameter in a bulk crystal, $\beta = 0°$, $\theta = 24°$, $\Gamma L = 2.7$, $a = 64$ •: experimental data, solid line: curve predicted by the theory including the nonlinear response empirical correction function, dashed–dot line: standard theory curve for the value of $\Gamma L = 0.43$, (value expected from the standard theory) dashed line: standard theory curve for $\Gamma L = 3$.

We have used the second order perturbation model to compare with our experimental data and obtained a good quantitative agreement. Figs. 2 and 3 show examples of the experimental data obtained in the bulk crystal, for beams focussed to spot sizes of $2w_0 \simeq 400\,\mu m$ and $2w_0 \simeq 100\,\mu m$, respectively.

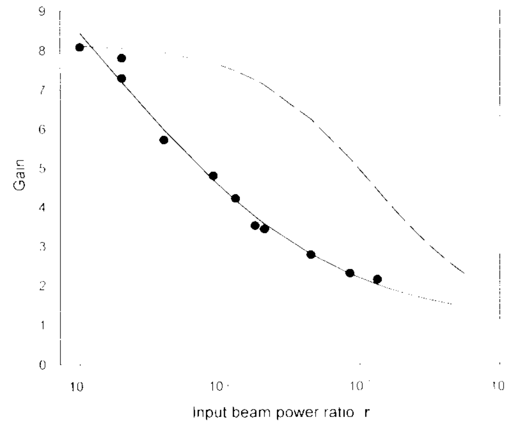

Fig. 4. Two-beam coupling gain as a function of the input power ratio for the case of beams focussed to $2w_0 = 10\ \mu m$ in diameter in a waveguide $\beta = 0°$, $\theta = 24°$, $\Gamma L = 2.13$, $a = 4.3$ •: experimental data, solid line: curve predicted by the theory including the nonlinear response empirical correction function, dashed–dot line: standard theory curve for the value of $\Gamma L = 0.013$, (value expected from the standard theory) dashed line: standard theory curve for $\Gamma L = 2.1$.

Fig. 4 presents data obtained in a waveguide, i.e. with the beams focussed to approximately $10\ \mu m$. Theoretical fit, using Eq. (11), and shown as solid lines, approximates the data quite well, but requires this enhanced value of coupling coefficient Γ_{new} with associated fitting parameter a. For the examples given in the three Figs. 2, 3, and 4, $\Gamma_{new}L$ was equal to 3, 2.7 and 2.1, and a to 11, 64, and 4.3, respectively.

The new coupling coefficient Γ_{new} is defined as [25]:

$$\Gamma_{new} = \frac{2\pi}{\lambda \cos\theta}\, r_{eff}\, n_0^3\, \frac{f(m)}{m}\, E_{w_G}, \qquad (12)$$

where E_{w_G} represents the magnitude of the space-charge field, which differs from the standard field induced by two plane (or Gaussian) waves.

To visualise better the anomalous behaviour of the experimentally determined gain, the standard plane waves two-beam coupling theory is also plotted for two values of ΓL. One curve is for the value of ΓL expected from the experimental geometry and the standard plane wave theory, and the second curve is the best fit using the standard theory with ΓL as a fitting parameter. As can be seen in Fig. 3, the difference between the experimental and the standard theory fit curves reaches up to three and a half orders of magnitude. This difference is in fact greater that the offset observed for two-beam coupling in a waveguide, as can be seen from a comparison with Fig. 4.

We suggest that the high value of coupling coefficient Γ_{new} arises from the non-uniform structure of the space charge field, and we propose the following processes as a qualitative explanation of its effect on the transport of carriers, and subsequently on beam coupling.

Firstly, the Gaussian distribution of the intensity pattern, with a sharp envelope in the extreme case, forms a strong, transverse spatial illumination gradient. This spatial pattern is reflected in the modulation of the space charge field, and therefore the steep maxima of the field affect the directional flow of photocarriers. The net result of this effect can be regarded as similar to an additional, very strong internal field. The presence and the origin of internal currents in $BaTiO_3$ has already been a point of some controversy; the bulk photovoltaic current is normally neglected in $BaTiO_3$, but it appears along the c axis of the crystal [31]. The kinetics of the photovoltaic field depend not only on the intensity distribution of the irradiating light, but also on the temperature.

In general, thermal effects can change various material parameters, such as refractive index, electro-optic coefficient or dielectric constant. Additionally, we also have to consider the possible effect of the thermal nonequilibrium created by the focussed beams, which may have a peak intensity approaching 20 kW/cm^2 in the most tightly focussed case. In fact, anomalously high diffraction has been observed in crystals where the formation of the grating was due to highly mobile nonthermalized electrons [32]. If the energy of carriers is much higher than the kinetic energy of the sample at room temperature (k_BT) then the diffusion field E_D, and hence, the gain, is governed by this energy excess $\Delta\varepsilon$ before the photocarriers thermalise. In experiments it has been found that the high mobility of nonthermalised charges can increase the diffusion field by 25 times with respect to the equilibrium diffusion value. Moreover, high intensity of input beams can change the effective trap concentration, and therefore the magnitude of the space charge field.

Currently, it is difficult to estimate the relative contribution of each of these effects on the magnitude and modulation of the space charge field, and hence the size of the coupling coefficient cannot be calculated exactly. We expect that to obtain the full picture of the TBC process with focussed beams, several geometrical and physical factors must be taken into account. Gaussian profiles of the interacting beams, plus the quantitative model of the enhanced mechanism of space-charge field, together with its nonlinear modulation would have to be solved simultaneously. Therefore, further research is necessary to investigate in more detail the exact origin of these effects.

6. Conclusions

We have carried out a detailed study of the two-beam coupling interactions with different degrees of beams focusing. The anomalous, previously unreported high gain has been measured showing a dependence on the input power ratio that is not typical.

We have shown that these effects cannot be explained on the basis of existing models which account for various intensity dependent photorefractive properties, and different spatial profiles of interacting beams. We have incorporated into the theory the empirical function to account for the modified space charge field, which we suppose may be enhanced and modified due to the strong illumination gradient, and behave similarly as under non-stationary conditions. The theoretically calculated gain dependence on the input power agreed, with reasonable accuracy, with the experimentally determined dependence. Little is still known about the exact mechanism that causes the increase of value of the coupling coefficient, but we suggest that the steep gradient of the focussed Gaussian beam profile of the space charge field acting on charges as an additional internal field could contribute to it. Also thermal effects, including the non-equilibrium, dominant diffusion of carriers, previously used to explain strong photorefractive response in other crystals, could also be responsible for the space–charge field enhancement. This first measurement and introductory modelling of the anomalous high gain, together with its fast build-up time, may prove very useful in the search for optimum operating conditions to achieve high efficiency, large sensitivity and fast response in photorefractive materials.

Acknowledgements

We would like to thank Daniel Fluck for his contribution to this work by establishing the effect of Gaussian profiles, and George Brost, Laszlo Solymar and George Valley for a number of interesting ideas and fruitful discussions. We gratefully acknowledge Marvin Klein and Barry Wechsler for supply of $BaTiO_3$ wafers and collaborative help. Also, thanks to Stephen James, Philip Jeffrey for initial experiments which revealed these non-standard beam coupling results.

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