

$+ A_s \exp(-j[k_s x - \omega_s t]) + \text{constant}$  (3)

Indexing terms: Optical fibres, Harmonic generation

A mechanism for phase-matched generation of second-harmonic (SH) light in an optical fibre has been experimentally and theoretically studied. A weak non-phase-matched SH signal, created via the quadrupole polarisation, initiates the self-writing of an axially periodic pattern of colour centres which is postulated to lead to the growth of a  $\chi^{(2)}$  grating. The grating (at the phase-match period) gives rise to SH generation at efficiencies of 0.5%. The spectral bandwidth of this  $\chi^{(2)}$  grating is 0.24 nm.

For exact phase-matching between  $E_p$  and  $E_s$ ,  $K = 2k_p - k_s$ , with  $k_p = (2\pi/\lambda_p)N_p$  and  $k_s = (2\pi/\lambda_s)N_s$ , where  $\lambda_p$ ,  $\lambda_s$  are vacuum wavelengths and  $N_p$ ,  $N_s$  are the effective refractive indices. Putting eqn. 3 into the wave equation with  $P$  as polarisation, neglecting second-order spatial derivations of the amplitudes  $A_i$  and placing the coefficients of terms with identical phase velocities to zero, leads to the following pair of equations:

**Introduction:** Second-harmonic generation (SHG) by an electric-dipole process is normally forbidden in a fused-silica fibre for two reasons: (i) silica possesses a centrosymmetric structure and (ii) it is difficult to achieve a phase-matching condition. Despite this, however, SHG of an Nd:YAG laser operating at 1.06  $\mu\text{m}$  has been recently reported<sup>1</sup> with efficiencies over 1%. The second-harmonic (SH) signal was observed to grow exponentially over a period of 8 h. We report here similar efficiencies for SHG, together with experimental evidence that the effect is due to the existence of a spatially periodic 2nd-order nonlinear susceptibility  $\chi^{(2)}$  in the fibre. Our hypotheses and experimental results are further supported by a coupled-wave theory in which it is shown that the phase-matching condition can be satisfied by the  $\chi^{(2)}$  grating vector.

$$\left. \begin{aligned} \frac{da_p}{dx} + j\gamma a_p^* a_s &= 0 \\ \frac{da_s}{dx} + j2\gamma^* a_p^2 &= 0 \end{aligned} \right\} \quad (4)$$

$$\gamma = \kappa^{(2)} \{ \exp[-jv^{(2)}x] + 0.5M^{(2)} \exp[jv_p x] \} \quad (5)$$

where

$$\left. \begin{aligned} a_i &= A_i/A_p(0) \\ v^{(2)} &= k_s - 2k_p \\ v_p &= (\Delta\lambda_p/\lambda_{po})n^{(2)} \\ \kappa^{(2)} &= \frac{A_p(0)k_p\chi^{(2)}}{N_p^2} \end{aligned} \right\} \quad (6)$$

are, respectively, the field amplitudes normalised to the initial pump amplitude  $A_p(0)$ , the SH dephasing parameter, the grating dephasing parameter and the SH coupling constant. Power conversion is obeyed by solutions of eqns. 4 since one can show that  $|a_s|^2 + 2|a_p|^2$  is constant. The key to the very large enhancement in SHG produced by a  $\chi^{(2)}$  grating lies in eqn. 5. The first part of  $\gamma$  yields the usual highly dephased (and hence very inefficient) SH coupling for constant  $\chi^{(2)}$ . The second part contributes SH coupling that is exactly phase-matched if  $v_p = 0$ , i.e. if  $\lambda_p = \lambda_{po}$ . Neglecting therefore the first term in  $\gamma$ , and solving first for zero pump depletion ( $da_p/dx = 0$ ):

**Physical mechanism:** In silica optical fibres very weak SHG occurs at high pump intensities owing to a nonlinear electric quadrupole susceptibility.<sup>2</sup> For a pump wavelength  $\lambda_p$  of 1.064  $\mu\text{m}$  the coherence length  $\Lambda_c$  between the pump and the SH at 532 nm is approximately 30  $\mu\text{m}$  as a result of both material and waveguide dispersion. This gives rise to a 30  $\mu\text{m}$  periodic intensity pattern of SHG (green) light along the fibre as energy is interchanged between pump and SH generated light.

In certain fibres with  $\text{SiO}_2$  cores doped with  $\text{GeO}_2$  and  $\text{P}_2\text{O}_5$ , semipermanent colour centres can be created by short-wavelength radiation.<sup>3</sup> These are known to form dipole centres which we postulate give rise to enhanced second-order nonlinear susceptibility. If this is the case, phase matching will be automatically achieved because the colour centres which enhance  $\chi^{(2)}$  will be written periodically into the fibre at spatial locations where the green light intensity is highest, i.e. where the pump and second-harmonic signal are in phase. At points where the pump and SH are out of phase few colour centres are generated. This results in reduced  $\chi^{(2)}$  and hence no destructive interference between existing generated green light and the second-order polarisation due to the nonlinear process.

The formation of colour centres is slow at 532 nm and increases rapidly with intensity. Thus it takes time to create the spatial grating in  $\chi^{(2)}$  within the fibre, and this gives rise to the observed rapid growth of SH light.

**Coupled-wave analysis:** We assume that the self-written  $\chi^{(2)}$  is sinusoidally modulated with distance  $x$  along the fibre at a period equal to the coherence length  $\Lambda_c$  of the SH polarisation at  $\lambda_p = \lambda_{po}$  (the writing wavelength). This gives an electric polarisation

$$P = \epsilon_0(E_p\chi_p + E_s\chi_s) + \epsilon_0\chi^{(2)}[1 + M^{(2)}\cos Kx]E^2 \quad (1)$$

where

$$E = E_p + E_s \quad (2)$$

$K = 2\pi/\Lambda_c$ ,  $M^{(2)}$  is the modulation strength of the grating,  $\chi_p$  and  $\chi_s$  are the susceptibilities at the pump ( $\omega_p$ ) and SH ( $\omega_s$ ) frequencies, and  $E_p$  and  $E_s$  are the corresponding electric fields. As we shall show, this  $\chi^{(2)}$  grating automatically ensures phase-matching (at  $\lambda_p = \lambda_{po}$ ) between  $E_p$  and  $E_s$ . The resulting total electric field is

$$|a_s|^2 \quad (x \leq L) = \{ \kappa^{(2)} M^{(2)} x \}^2 \text{sinc}^2 \{ v_p x / 2 \} \quad (7)$$

where  $L$  is the total grating length. Hence for conversion efficiencies of less than about 5% the spectral bandwidth  $\Delta\lambda_{\mu(1/2)}$  of the SHG process is

$$\Delta\lambda_{\mu(1/2)} = 4\sqrt{(2)\lambda_{po}/v^{(2)}L} \quad (8)$$

In the presence of pump depletion and for  $v_p = 0$  an exact solution exists:

$$|a_s|^2 \quad (x \leq L) = 2 \tanh^2 \{ \kappa^{(2)} M^{(2)} x / \sqrt{2} \} \quad (9)$$

Thus peak conversion efficiency to the SH is approximately quadratic with  $x$  for  $\kappa^{(2)} M^{(2)} x \ll 1$ . However, much more significant is that the  $\chi^{(2)}$  grating potentially yields efficiencies approaching 100% for large enough  $L$ , as is commensurate with phase-matching.

**Experiment:** Mode-locked, Q-switched pulses of <100 ps duration and peak powers of 10 kW from an Nd:YAG laser at 1.064  $\mu\text{m}$  were launched into a variety of single-mode fibres. Very weak second- and third-harmonic generation was observed in several. To date, however, growth in SHG has only been observed in a silica fibre with the core and cladding doped with <1%  $\text{P}_2\text{O}_5$  and the core doped with 15%  $\text{GeO}_2$ . The SH signal grew with time from 100 pW to 50 W over a period of 10 h.

To confirm the establishment of a  $\chi^{(2)}$  grating, the wavelength dependence of SHG was measured. Pulses from a tunable Raman-shifted dye laser of 6 ns duration and peak powers of 1 kW were launched into a fibre with a previously burnt-in  $\chi^{(2)}$  grating. The pump was tuned over 4 nm, giving a measured  $\Delta\lambda_{p(1/2)}$  of 0.24 nm for the SH signal, as shown by the experimental points in Fig. 1. The same behaviour resulted when the fibre was pumped from the other end. A best fit of eqn. 4 to the experimental data occurs at an effective grating length  $L$  of 12 cm, with  $\nu^{(2)} = 2\pi/\Lambda_c = 0.207 \mu\text{m}^{-1}$ . Side-scatter measurements nearer the input end of the grating region gave significantly broader linewidths, as one would expect from eqn. 8.

Fig. 1 shows that the largest discrepancies between experiment and theory occur in the sidelobes. This is likely to be linked to nonuniformities in grating phase (and to a lesser extent  $M^{(2)}$  and  $\chi^{(2)}$ ) along the fibre. Such variations are an inevitable consequence of the self-writing process, and a more elaborate theory is at present being developed to deal with these effects. Their presence is confirmed in Fig. 2, where a side-scatter measurement of the SH intensity is plotted as a function of distance from the fibre end. The pump light was launched from the opposite end of the fibre. It can be appreciated that about 90% of the conversion occurs in the last 15 cm, which agrees quite well with our best-fit value of 12 cm.

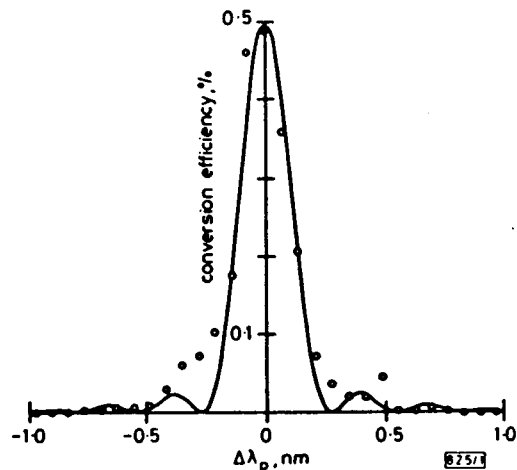


Fig. 1 Wavelength dependence of SH conversion for deviations of pump wavelength from 1.064  $\mu\text{m}$

Circles are experimental points and solid curve a best theoretical fit

A variety of different phenomena are likely to contribute to these axial nonuniformities in grating properties, such as the manifestly nonlinear  $x$ -dependence of the growing SH signal, group velocity walk-off between pump and SH, and pump depletion due to stimulated Raman or Brillouin scattering. The efficiency of SHG—as distinct from the writing process—will level off at very high pump powers owing to competing nonlinear processes such as the two just mentioned.

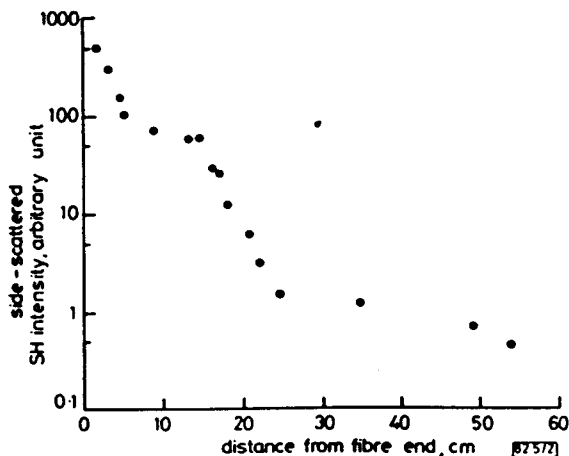


Fig. 2 Measured side-scattered SH intensity as a function of distance from fibre end

While the  $\chi^{(2)}$  grating was written with pump light launched from the left, this measurement was made with pump light launched from the opposite (RH) end

Finally, an estimate of the product  $\chi^{(2)}M^{(2)}$ , the modulation depth of  $\chi^{(2)}$ , can be made if we take our result of 0.5% conversion efficiency at  $\lambda_p = \lambda_{ps}$  and compare it with the peak value given by eqn. 7. For the fibre spot-size diameter of 5  $\mu\text{m}$ ,  $\lambda_{ps} = 1.064 \mu\text{m}$  and a peak pulse power of 10 kW, 0.5% conversion occurs if  $\chi^{(2)}M^{(2)} = 2.75 \times 10^{-16} \text{ m/V}$ . From eqn. 9, 60% conversion would be achieved if a grating of this strength were uniform over 1.7 m of fibre.

**Conclusions:** We have confirmed experimentally that 1% efficient second-harmonic generation is possible in an optical fibre. We postulate that the mechanism is due to the generation of a grating of colour centres which enhance the second-order susceptibility. The grating period is matched to the coherence length between pump and SH waves. Confirmation of the grating model is provided by our measurements of the operating bandwidth as 0.24 nm.

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M. C. FARRIES  
P. ST. J. RUSSELL  
M. E. FERMANN  
D. N. PAYNE

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Optical Fibre Group  
Department of Electronics & Computer Science  
University of Southampton  
Southampton SO9 5NH, United Kingdom

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