

GENERATION OF PERMANENT OPTICALLY-INDUCED 2ND ORDER  
NONLINEARITIES IN OPTICAL FIBERS BY POLING

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**Abstract**

It is shown that large permanent enhancements in second-order optical nonlinearity can be induced in germanosilicate fibers (both pure and co-doped with P) by application of a transverse dc electric poling field in the presence of high intensity light. The macroscopic inversion symmetry of the core material is broken by the excitation and alignment of defect centers. Significant frequency doubling results despite the absence of phase-matching. The saturation (both with increasing dc-field and intensity) of this effect is investigated.

The amorphous nature of glasses normally disallows the presence of any second-order optical nonlinearity through lack of macroscopic non-inversion symmetry. Hence considerable interest has been generated by the observation of Österberg and Margulis<sup>1</sup>, that anomalously efficient second-harmonic (SH) generation can appear in optical fibers exposed to intense light at 1.064  $\mu\text{m}$ . There is now fair agreement over how this phenomenon arises; phase-matching is achieved via a periodically modulated  $\chi^{(2)}$  whose grating period equals twice the length over which the frequency-doubled infra-red polarization and the SH light get 180° out-of-phase<sup>2</sup>; and non-inversion symmetry arises through the optical mixing via  $\chi^{(3)}$  of two pump photons and one SH photon<sup>3</sup>. The resulting dc polarization reverses sign at the correct period for phase-matching.

The final link in the argument is that this dc polarization causes (in the presence of intense light) the alignment or realignment of defects, whose dipole moments then create a  $\chi^{(2)}$  grating. Stolen and Tom<sup>3</sup> showed that a fiber could be rapidly prepared for SH generation by launching 1.064  $\mu\text{m}$  and 0.532  $\mu\text{m}$  light simultaneously into the fiber. This provided confirmation that the model is essentially correct. However there remains one outstanding difficulty; the dc polarization field is tiny. Taking  $\chi^{(3)} = 10^{-22} (\text{m/V})^2$ , infra-red (IR) and SH powers of 1 kW and 40 W, and a core diameter of 3  $\mu\text{m}$ , it turns out to be around 4 V/cm. Can so small an electric field have so profound an effect, and if so, surely one could obtain much greater enhancements in  $\chi^{(2)}$  by applying a large external electric field? It has recently been

shown that  $\text{LiNbO}_3$  and  $\text{LiTaO}_3$  crystals can be poled using an electron beam as excitation source<sup>4</sup>.

An obvious way of resolving these questions is to apply an external field in the presence of intense light using systems of in-built capillary electrodes<sup>5</sup>. It is also of interest to investigate the effect of phosphorus as co-dopant, since it has recently been shown that phosphorus is not essential for observation of enhanced SH generation<sup>6</sup>. To this end we fabricated two fibers with different geometries and compositions (Figure 1). The first (fiber GS) had a pure germanosilicate core (28 mole%  $\text{GeO}_2$ ) of diameter 10  $\mu\text{m}$ , and the second (fiber PGS) a P-doped germanosilicate core (13 mole%  $\text{GeO}_2$  with 0.5 mole%  $\text{P}_2\text{O}_5$ ) of diameter 3.2  $\mu\text{m}$ . Fiber GS had two capillary electrodes on opposite sides of the core, and fiber PGS a D-shaped cross-section with one external and one internal electrode. The maximum fields we could apply before breakdown were limited to 20  $\text{V}/\mu\text{m}$  in fiber GS and 100  $\text{V}/\mu\text{m}$  in fiber PGS. Fiber PGS was weakly polarisation preserving. Two different lasers were used to excite defect centers for alignment (or realignment) in the applied dc field. These were 1) a pulsed dye-laser operating at 485 nm (6 nsec pulse width and 30 Hz repetition rate) and 2) a CW Argon laser operating at 488 nm. We chose these wavelengths because of our suspicion that two or three-photon effects (at 1.064  $\mu\text{m}$ ) are the cause of defect excitation in the original observations of SHG in fibers<sup>7</sup>. Blue light is also known to be much more efficient than longer wavelengths at exciting color-centers in germanosilicate fibers<sup>8</sup>.

We were able to estimate the permanent enhancement in  $\chi^{(2)}$  caused by defect alignment as follows. It is well known that a temporary second-order nonlinearity can be induced in almost any material by applying a transverse dc electric field. The resulting effective second-order nonlinearity  $\chi_{dc}^{(2)}$  is given by:

$$\chi_{dc}^{(2)} = \chi_{1111}^{(3)} E_{dc}$$

So, before launching any blue light into the fiber, we applied a dc field  $E_{dc}$  and coupled in a low level of IR light. The resulting SH power  $P_{dc}^{2\omega}$  allowed us to establish a reference level of second-order nonlinearity against which any subsequent enhancement due to defect alignment could be estimated. Assuming the phase mismatch to be unaffected by the alignment process, the following expression results for the enhanced second-order nonlinearity  $\chi_{enh}^{(2)}$ :

$$\chi_{enh}^{(2)} = E_{dc} \chi_{1111}^{(3)} \left[ P_{enh}^{2\omega} / P_{dc}^{2\omega} \right]^{1/2} \quad (1)$$

where  $P_{enh}^{2\omega}$  is the SH power (for the same low level of IR power) measured after switching off the dc field and blocking the blue light. Instabilities in peak IR power meant that  $\chi_{enh}^{(2)}$  as estimated by Equation (2) was accurate to  $\pm 20\%$ .

Prior to any treatment of the fibers, a very weak SH signal (caused by non-linear quadrupole effects and magnetic dipole moments<sup>9</sup>) was detected. A dc field was then applied across the fiber, and blue light launched into it. At regular intervals the

blue light was blocked and the dc field switched off, and the growth in  $\chi^{(2)}$  monitored by launching pulses at  $1.064 \mu\text{m}$  into the fiber and measuring the SH conversion efficiency. The peak IR pulse power was limited to 75 W so as not to disturb the aligned defect centres.

1) Pulsed blue light at 485 nm as excitation source

A minimum in-core peak pulse intensity  $I_{th}$  of  $1 \text{ W}/\mu\text{m}^2$  was necessary before any increase in SH signal was detectable. At intensities above this threshold the SH conversion efficiency typically took several minutes to reach a saturation value  $P_{sat}^{2\omega}$  that depended on  $E_{dc}$ . The maximum observed SH power in fibre GS at  $E_{dc} = 4 \text{ V}/\mu\text{m}$  is plotted in Figure 2 as a function of peak blue light intensity. Just above  $I_{th}$  this function is strongly nonlinear, and for higher intensities it saturates. Once created, the induced  $\chi^{(2)}$  non-linearity was permanent. It could not be erased by blue light alone, even at intensities higher than those used in the alignment process. This suggests that cooperative effects between aligned defects, leading to enhanced orientational stability, may play a significant role in the phenomenon.

The saturated SH power  $P_{sat}^{2\omega}$  in fiber GS at a constant peak blue pulse intensity of  $3.2 \text{ W}/\mu\text{m}^2$  is plotted as a function of dc-field strength in Figure 3. For small fields  $P_{sat}^{2\omega}$  grows quadratically with electric field, and above about  $4 \text{ V}/\mu\text{m}$  it saturates. This suggests that almost perfect alignment of all the available orientable defect centres is achieved for  $E_{dc} > 4 \text{ V}/\mu\text{m}$ . From these results we estimate from Equation 1 that a

maximum enhanced  $\chi^{(2)}$  of  $3 \times 10^{-15}$  m/V is possible in fibre GS using a pulsed blue laser as the defect excitation source.

The same experiment for the P-doped fiber PGS yielded a maximum enhanced  $\chi^{(2)}$  of  $3 \times 10^{-15}$  m/V for the same in-core blue light intensity but with an applied dc-field of  $100 \text{ V}/\mu\text{m}$ . This indicates that, for pulsed blue light excitation, the P-doping does not lead to an increase in  $\chi^{(2)}$  and that no advantage is gained by using dc-fields higher than about  $20 \text{ V}/\mu\text{m}$  in the alignment process.

## 2) CW light at 488 nm as excitation source

Argon laser light at 488 nm was launched into fibres GS and PGS while orientating fields of respectively  $15 \text{ V}/\mu\text{m}$  and  $100 \text{ V}/\mu\text{m}$  were applied. The irradiation time was 10 minutes and the in-core intensities  $10 \text{ mW}/\mu\text{m}^2$  (fiber GS) and  $50 \text{ mW}/\mu\text{m}^2$  (fiber PGS). The estimated enhanced  $\chi^{(2)}$  for fiber GS was  $3 \times 10^{-15} \text{ m/V}$ . For fiber PGS it was  $7 \times 10^{-14} \text{ m/V}$ , which is some 20x larger than achieved with pulsed blue light. This shows that large average blue light powers are more efficient at exciting defect centres in fiber PGS than blue pulses of low average power. However, the factor of difference is much smaller than the  $10^6 \times$  increase in average power (and  $500 \times$  reduction in peak power) would suggest. This may indicate that both single-photon and two-photon absorption play a role.

## Discussion

The level of second order nonlinearity obtained with fiber PGS under CW excitation ( $7 \times 10^{-14}$  m/V) is more than 100 times larger than achieved by relying on the internal dc polarisation for alignment<sup>2,3</sup>. The conversion efficiencies we obtain (0.001%

in fibre PGS at an infrared power of 300 W) are smaller only because of the lack of a good phase-match. Significantly higher conversion efficiencies should be possible by applying a transverse dc alignment field that reverses sign periodically with distance along the fiber axis. This would create a phase-matching structure<sup>2</sup>.

We have also made some preliminary measurements using red and infra-red light to excite the defect centers in the presence of an aligning field. Some enhancement in SH signal resulted, although much higher power levels were needed to create second-order nonlinearities of magnitudes similar to the ones obtained with blue light. We are at present looking in more detail at the wavelength dependence of defect excitation. The orientation of defect centres in an external dc electric field should also be possible in bulk glasses and may provide a suitable method for assessing the usefulness of different glasses for SH generation. The technique also offers a means of establishing the form of the  $\chi^{(2)}$  tensor more precisely, a measurement that has previously been limited by birefringence and imperfections in optical fibers<sup>10</sup>.

In conclusion, an external dc poling field has a similar effect on defects as the internal polarization proposed by Stolen and Tom<sup>3</sup>. In the presence of blue light, defects are excited and then re-align with the dc field. Since the external fields can be up to 5 orders of magnitude larger than the polarization, the alignment process is much more effective. Saturation mechanisms limit the maximum induced second-order nonlinearity. The induced  $\chi^{(2)}$  cannot be erased by blue light alone, which suggests that

cooperative effects between aligned defects enhance their stability of alignment.

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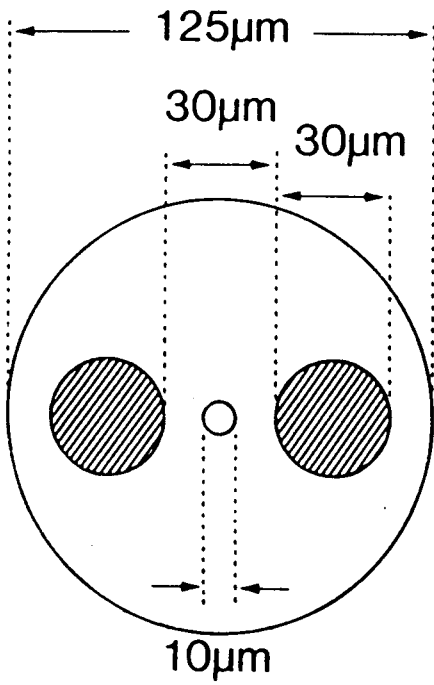
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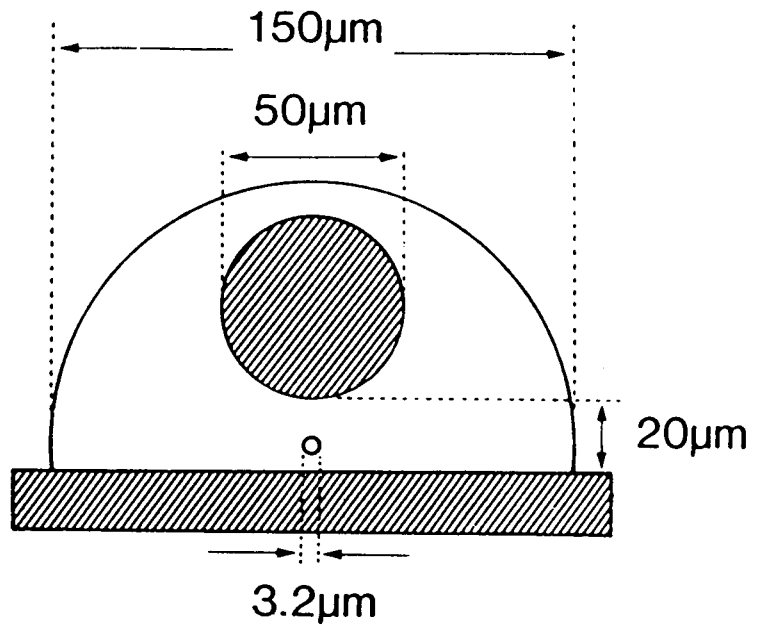
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## FIGURE CAPTIONS

1. Cross-sections of fibers used in the experiments. Fiber GS is pure germanosilicate, and fiber PGS P-doped. The shaded areas indicate the metal electrodes. A plane metal plate was pressed externally against the side of the D-shaped fiber PGS to provide the second electrode.
2. Saturated conversion efficiency to the SH in fiber GS as a function of launched peak pulse intensity (at 485 nm) at a constant dc alignment field of  $4 \text{ V}/\mu\text{m}$ .
3. Enhanced SH-power as a function of applied dc electric field strength in fiber GS for a constant launched peak pulse intensity (at 485 nm) of  $3.2 \text{ W}/\mu\text{m}^2$ .



Fibre GS



Fibre PGS

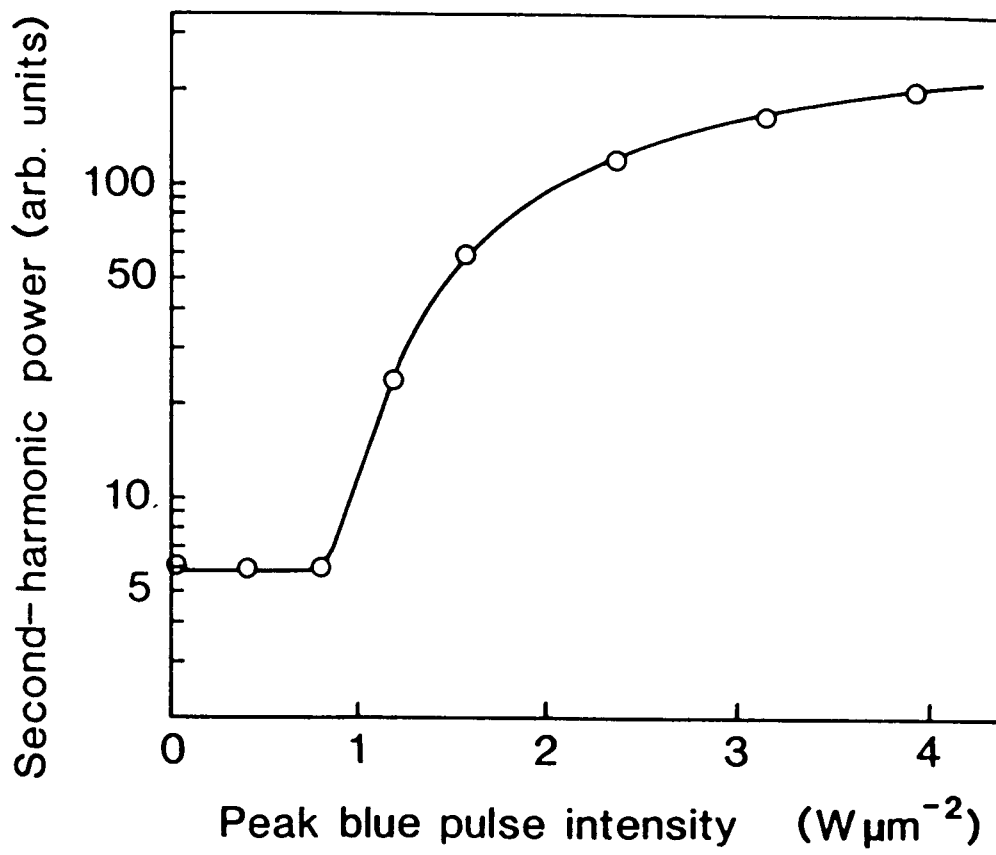


Figure 2

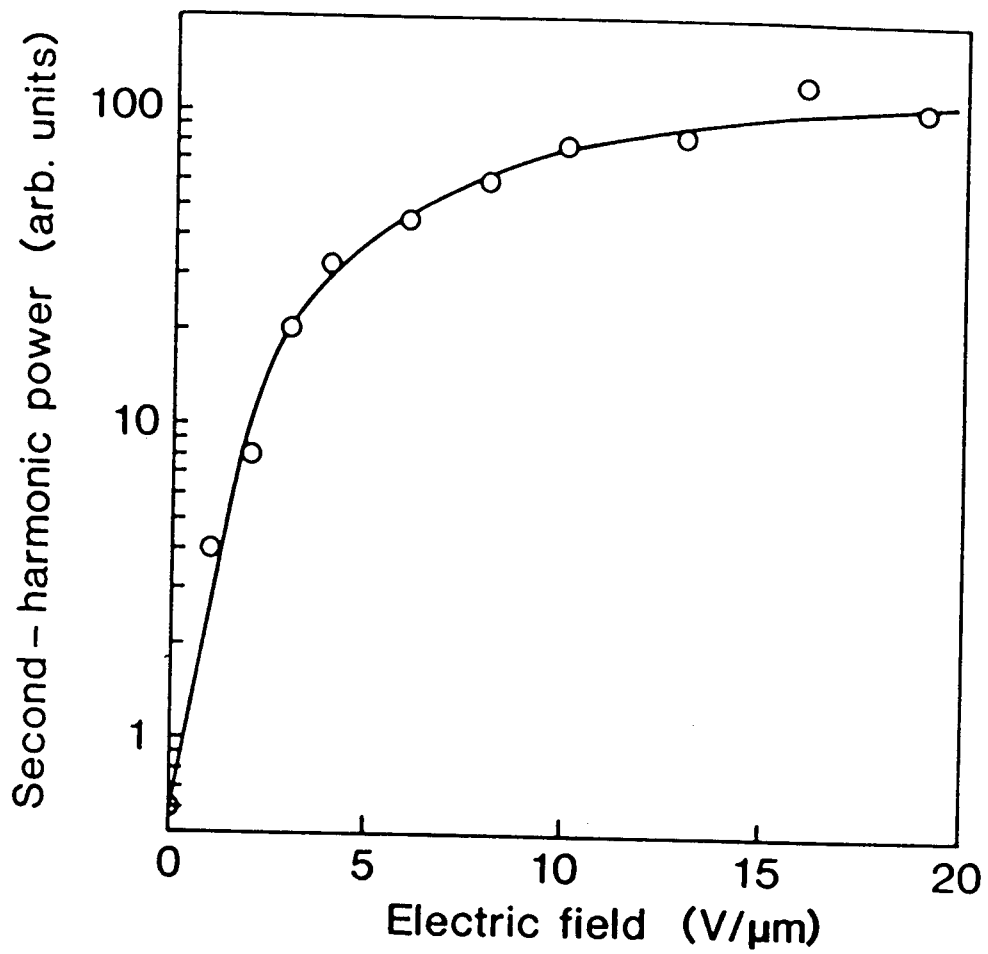


Figure 3