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Impurity enhanced self-pumped phase conjugation in the near infrared in 'blue' $BaTiO_3$

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Photorefractive self-pumped phase-conjugation is examined at near-infrared wavelengths using doped 'blue' $BaTiO_3$, and reflectivities are reported as high as 76% between 720 nm and 1004 nm due, as is believed, to a backward stimulated photorefractive scattering mechanism. Data also suggests that it is possible to stabilise the phase-conjugate reflectivity by vibrating the crystal.

Since the discovery of total-internal-reflection self-pumped phase-conjugation (TIR SPPC) using photorefractive $BaTiO_3$ [1], observation of the effect has been carried out mainly in the visible part of the spectrum where crystal response has tended to be most efficient. However, with the availability of diode lasers operating at near infrared wavelengths, attention has been drawn to the possibility of extending the crystal response to allow observation of TIR SPPC and other photorefractive processes at diode compatible wavelengths. Recently, efficient TIR SPPC using a nominally undoped crystal has been observed with reflectivities as high as 72% at 800 nm [2], and there has been a concerted effort to extend and enhance the response of $BaTiO_3$ in the near infrared region via the addition of different dopants. The effect of cobalt as a dopant is discussed in ref. [3], for which a reflectivity of 50% at 933 nm was reported. To our knowledge this is the longest wavelength to date at which TIR SPPC has been observed using $BaTiO_3$ operating at room temperature, and without the application of any external electric fields. Phase conjugation has been previously reported at 1.06 μm (with a reflectivity of ~30%) [4] and at 1.09 μm (with a reflectivity of 18%) [5], but both these experiments involved a ring configuration requiring additional optics. In this paper, however, we report high phase conjugate reflectivities of up to 76% out to 1 μm using an internal reflection geometry.

The $BaTiO_3$ crystal used in our experiments was grown by the top-seeded solution growth technique from 99.999% purity starting materials, annealed and electrically poled. An unknown accidental impurity produced an enhanced absorption in the red and near infrared with an absorption peak at about 640 nm that gave the crystal a blue colour. The absorption spectrum measurement (fig. 1) for this crystal was

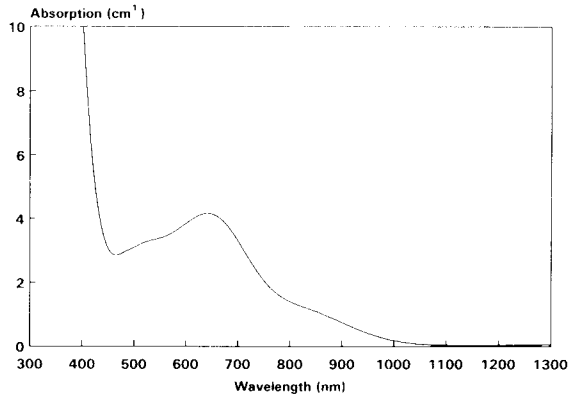


Fig. 1. Absorption spectrum of the 'blue' $BaTiO_3$ using light polarised parallel to the crystal c-axis.

performed with a Perkin–Elmer Lambda 9 spectrometer using light polarized parallel to the c-axis.

The crystal sample had dimensions $4.70 \times 2.69 \times 4.41$ mm^3 with the crystal c-axis parallel to the 4.41 mm edge. Using the experimental arrangement shown in fig. 2, the crystal, mounted in air on a combined rotation–translation stage, was placed in the standard TIR geometry with the beam incident upon the 4.41×2.69 mm^2 face, and a systematic study was undertaken to assess the phase conjugate reflectivity with respect to wavelength (λ), horizontal beam position on the entrance face (z), external angle of incidence (θ) and input power (I_{in}). The experimental parameters are shown in the inset in fig. 2. A Ti:sapphire laser pumped using a 5 W argon laser, and calibrated using an Anritsu optical spectrum analyser, was used to provide a tunable source of infrared radiation. The lens pair (L1 and L2, focal lengths 20 cm and -5 cm, respectively) served to collimate the beam and reduce the input beam diameter (ϕ) at the crystal to ~ 1 mm, and the polarising beam splitter (PBS) ensured that the beam was horizontally polarised. In fig. 2, I_0 (I'_0) is the intensity of the Ti:sapphire beam incident upon (reflected from) the beam splitter (BS), and I_{in}, is the intensity of the beam transmitted through the BS and incident upon the crystal. I_{pc} (I'_{pc}) is the intensity of the phase conjugate signal incident upon (reflected from) the BS. The phase conjugate reflectivity was then determined by measuring the ratio (I'_{pc}/I'_0) using a calibrated Newport power meter. Careful measurement of the beam splitter reflectivity allowed the phase conjugate reflectivity to be calculated by dividing this ratio by $(1-R)^2$, where R is the reflectivity of the beam splitter. Note that the reflectivities reported in this letter are not corrected for Fresnel reflections at the crystal, but instead represent the actual fraction of input power reflected and, therefore, indicate a lower bound for the phase conjugate reflectivity. The parameters for each experiment are listed in the figure captions.

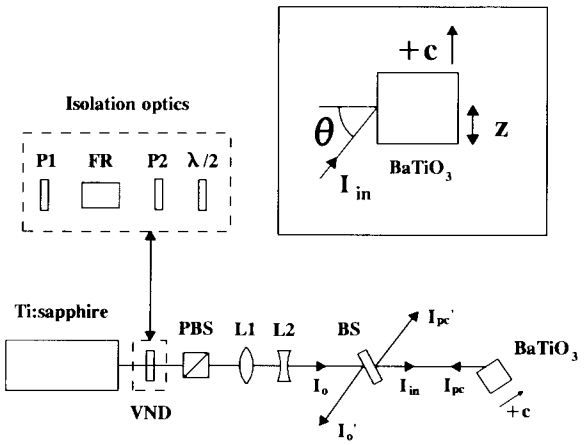
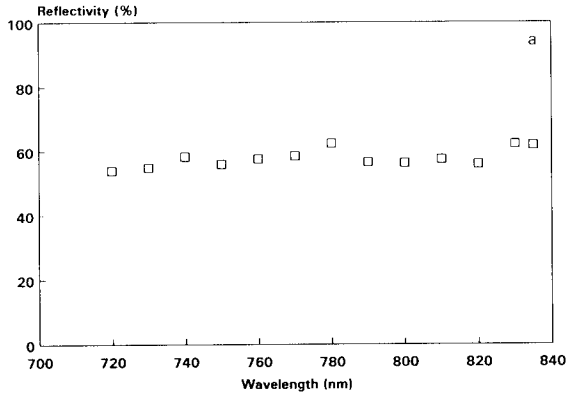


Fig. 2. Experimental arrangement. P1, P2, polarisers; FR, Faraday rotator; $\lambda/2$, half-wave plate. In order to measure response time against incident power, the VND was replaced by the isolation optics. Inset: the experimental parameters.

In determining SPPC behaviour in the wavelength range 720 nm to 835 nm, the initial incident power was adjusted to 45 mW using a variable neutral density filter (VND). The crystal was placed with $\theta = 60°$ and $z = 1.6$ mm. Results, shown in fig. 3a, demonstrate reflectivities of up to 62% which, however, was not an optimised value. At the longer wavelength range, 855 nm to 1004 nm (the longest wavelength available from the Ti:sapphire laser), fig. 3b, the phase conjugate reflectivity was optimised by removing the collimating lens pair, and instead placing the crystal downstream of a single 50 cm focal length lens to achieve $\phi \sim 0.5$ mm. The crystal itself was rotated to Brewster's angle ($\sim 67°$, as in ref. [2]) to both minimise the Fresnel reflection and also maximise access to the largest electrooptic coefficient. The VND was also removed in order to use all the available power from the Ti:sapphire laser. Monitoring I'_0 and I'_{pc} simultaneously revealed a rise in both signals as the phase conjugate formed, indicating that feedback from the phase conjugate mirror into the Ti:sapphire laser was causing a rise in incident intensity (I_{in}). At wavelengths corresponding to the extremes of the Ti:sapphire tuning range, this phase conjugate feedback caused a rise in the incident signal of up to a factor of 8, and at wavelengths closer to the centre of the tuning range, a rise of a factor of 2 was observed. Once the phase conjugate signal was established, however, the steady-state reflectivity was measured, using one power meter in order to avoid cross calibration problems. Reflectivities of up to 76% were observed. This feedback effect made measurement of response time against wavelength impractical without optical isolation at each wavelength.

During the course of the experiments, it was found



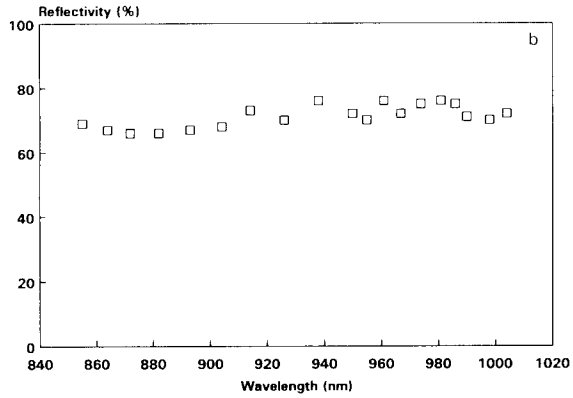


Fig. 3 (a). Graph of phase conjugate reflectivity against wavelength (λ). $\theta=60°$; $z=1.6$ mm; $\phi\sim 1$ mm; with instability suppression. (b) Graph of phase conjugate reflectivity against wavelength (λ). $\theta=67°$; $z=1.6$ mm; $\phi\sim 0.5$ mm; no stabilisation required.

that instabilities in the phase conjugate reflectivity could be removed by vibrating the crystal slightly. This effect was first realised when vibrations from an equipment cooling fan, placed close to the experiment on the optical table, appeared to stabilise the reflectivity. Figure 4 shows the improvement in stability when the vibration source was switched on at time t_0. The measurements shown in fig. 3a (with $\theta=60°$) were made using this vibration stabilisation, however in fig. 3b (with $\theta=67°$) no stabilisation was required. Further work is currently in progress to determine the exact cause of the improvement, but we suspect that the washing-out of competing, parasitic gratings allows stable and repeatable measurements.

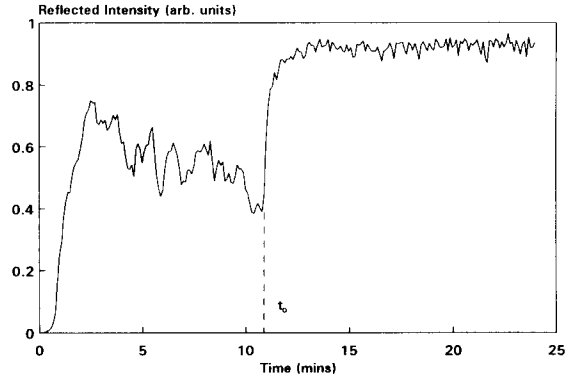


Fig. 4. Phase conjugate reflectivity behaviour with time illustrating instability suppression. $\phi=60°$; $z=1.6$ mm; $\lambda=800$ nm; $\phi\sim 1$ mm. Stabilisation was switched on at time t_0.

In order to further characterise the crystal, experiments to determine reflectivity with respect to z, θ and I_{in} were performed with the collimating lens pair in place, and instability suppression as outlined above. The Ti:sapphire was tuned to 800 nm and isolated by replacing the VND with the optics shown. Assessment of the crystal properties was begun by setting $\theta=60°$, and translating the crystal perpendicular to the incident beam, in order to monitor the phase conjugate reflectivity as a function of z (measured to the centre of the incident spot which had a projected horizontal diameter of ~ 2 mm). Results of this first analysis (fig. 5) show that SPPC in this particular geometry appears possible only over a small range with 1.0 mm $< z <$ 2.2 mm – the lower extreme of z coinciding with the incidence of a portion of the input beam on the $-c$ face of the crystal. With the entrance position fixed at $z=$ 1.6 mm, analysis of the behaviour of reflectivity with yields a curve (shown in fig. 6) which has a maximum reflectivity at an incident angle of $\sim 55°$. Examination of the behaviour of reflectivity with I_{in} was achieved by rotating the polarisation using the half wave plate, and rejecting the o-component at the PBS, and it was found that reflectivity ($\sim 63\%$) appears to remain independent of I_{in} over the range 7–70 mW. The response times over this range of I_{in} are shown in fig. 7 (note the logarithmic scales), where we define response time as the time taken for the phase conjugate signal to rise from 10% to 90% of its saturated value. After each measurement, the crystal was uni-

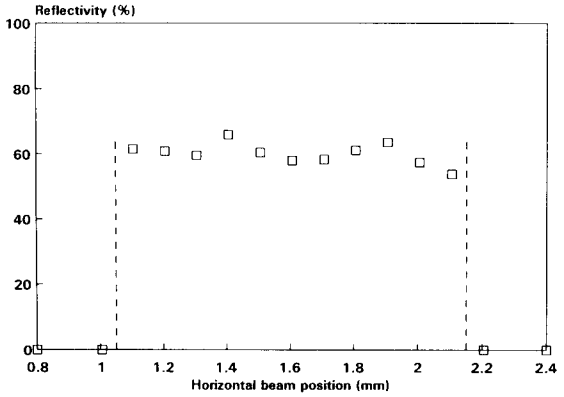


Fig. 5. Graph of phase conjugate reflectivity against horizontal beam position on crystal entrance face (z). $\theta=60°$; $\lambda=800$ nm; $\phi\sim 1$ mm; $I_{in}=45$ mW; with optical isolation and instability suppression. The dashed vertical lines indicate the range over which SPPC is observed.

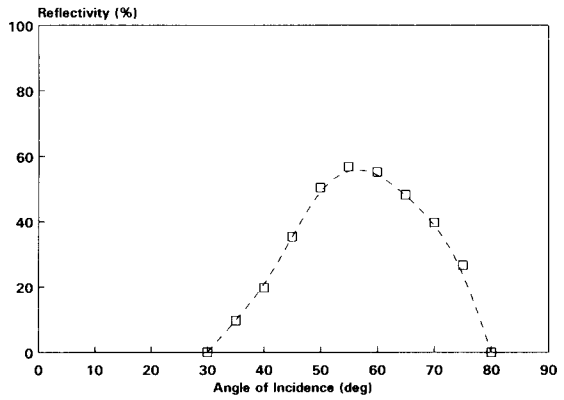


Fig. 6. Graph of phase conjugate reflectivity against angle of incidence of beam (θ). $z=1.6$ mm; $\lambda=800$ nm; $\phi\sim 1$ mm; $I_{in}=43$ mW with optical isolation and instability suppression. The dashed line is included as a guide for the eye.

formly illuminated for 2 minutes using a 75 W white light source to ensure that all gratings were erased. The straight line fit in fig. 7 would suggest that the response time is proportional to I_{in}^{-x} where $x=0.75$ – a sublinear relationship indicating, perhaps, the presence of shallow traps [6].

The unexpectedly high reflectivity (given the high crystal absorption) raises questions regarding the photorefractive mechanism responsible. If we assume the TIR (corner reflection) mechanism, and a round-trip path length of 1 cm for the beam travelling from the crystal entrance face to the TIR corner, then the absorption coefficient of $\alpha\sim 2$ cm^{-1} (from fig. 1) at 750 nm would imply that the maximum reflectivity possible at that wavelength is $e^{-2}=0.14$, i.e. 14% – a factor of four less than the measured reflectivity of 56% (see fig. 3a). This inconsistency between theoretical maximum (assuming the corner reflection geometry) and observed value would imply that the TIR mechanism assumption is, perhaps, inappropriate despite the crystal being placed in the TIR geometry. We believe that, in the absence of significant light induced transparency, reflection gratings near the entrance face of the crystal may perhaps be responsible for the high reflectivities by a backward scattering mechanism [7]. Further investigations are currently under way, as are investigations into the instability suppression outlined in this paper. As regards the SPPC, however, fig. 3b demonstrates that whilst stabilising the output at shorter wavelengths, vibrating the crystal is not, in fact, responsible for the high reflectivities obtained with this crystal.

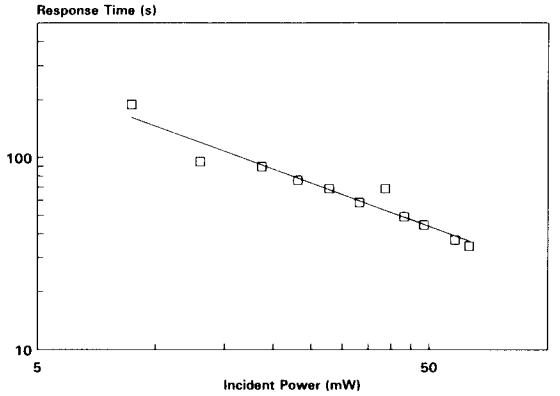


Fig. 7. Graph of SPPC response time against incident power (I_{in}). $\theta=60°$; $z=1.9$ mm; $\lambda=800$ nm; $\phi\sim 1$ mm; with optical isolation and instability suppression. The straight-line fit indicates an I_{in}^{-x} power law with $x=0.75$. Note that 10 mW incident power corresponds to ~ 0.64 W cm^{-2}.

In summary, we have carried out a systematic study of SPPC in the near infrared using a doped sample of 'blue' $BaTiO_3$ in the TIR geometry, and report highly efficient reflectivities of up to 76% at 1 μm, which we believe to be the highest reported to date.

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