

# PHOTOREFRACTIVE SCATTERING IN BULK PERIODICALLY POLED LITHIUM NIOBATE

V. Pruneri, P.G. Kazansky, J. Webjörn, P.St.J. Russell and D.C. Hanna  
Optoelectronics Research Centre, University of Southampton  
Southampton SO17 1BJ, U.K.

## *Abstract*

We report a new species of self-organised light-induced scattering, appearing only in bulk periodically poled lithium niobate. Clearly defined diffracted beams develop on exposure to 532 nm light from a frequency-doubled, Q-switched and mode-locked Nd:YAG laser. The structure thus created also diffracts light at 1064 nm. The effect is strongly dependent on temperature, the polarization state of the light and the period of domain reversal. It exhibits a sharply defined nonlinear threshold at which the far-field pattern of the diffracted light evolves rapidly.

## *Summary*

Lithium niobate displays a very strong photo-galvanic (sometimes known as photo-voltaic) effect [1] which, combined with the electrooptic effect, can produce significant changes in refractive index. This photorefractive effect, first observed nearly thirty years ago [2] has led to a number of applications. In nonlinear optical devices however, where preserving good beam quality is vital, it is an unwelcome effect, causing substantial beam distortion (photorefractive damage) in single-domain lithium niobate (SDLN).

Recently periodic poling (or domain inversion) has emerged as a reliable technique for achieving quasi-phase-matched (QPM) parametric frequency conversion. Domain inversion causes a reversal in the direction of the spontaneous polarization, the photo-galvanic current, and the sign of the electrooptic and nonlinear coefficients. Under the correct conditions, the reversal in the direction of the space charge field from domain to domain can lead to almost complete elimination of the photorefractive damage in periodically poled lithium niobate (PPLN). Experimental tests using green light from a CW argon ion laser [3,4] confirm that photorefractive damage is essentially absent in PPLN while being strongly present in SDLN. Although various different types of light-induced scattering due to photorefractive damage have been reported both in SDLN [5] and potassium niobate [6], this is the first time to our knowledge that similar phenomena have been experimentally observed in PPLN crystals.

Two samples of PPLN 0.2 mm thick and ~4 mm long were prepared with pitches (i.e. domain reversal periods) of 6.8  $\mu\text{m}$  (sample A) and 9  $\mu\text{m}$  (sample B). This periodic domain reversal was achieved using high-voltage pulses applied via liquid electrodes [3,7,8]. The measured nonlinear coefficient, via second harmonic generation, was for both samples ~70% of the theoretical value. This indicated that the geometrical quality of the periodic domain pattern was rather good.

Photorefractive damage was then investigated using the 488 nm line of a CW argon ion laser in the same way as described in [3], confirming that periodic domain inversion had essentially eliminated the photorefractive effect.

The experimental set-up for observing photoinduced scattering is shown in Fig.1.

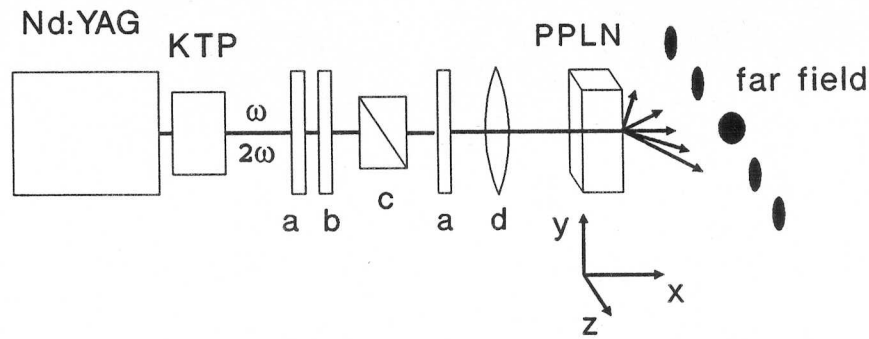


Fig.1 Experimental set-up: half-wave plate at 532 nm (a) and 1064 nm (b), polarizer (c), lens  $f=70$  mm (d).

The laser was Q-switched at a repetition rate of 1 kHz, each pulse consisting of a train of  $\sim 30$  mode-locked pulses (each separated by 10 ns) of  $\sim 300$  ps pulse duration across the  $\sim 300$  ns intensity FWHM of the Q-switched envelope. This pulse train was then frequency doubled to 532 nm in a KTP crystal. The beams were focused into the sample along its x-axis to a spot size of about  $40 \mu\text{m}$  for the green and  $50 \mu\text{m}$  for the infrared.

We first examined sample A. Photorefractive damage was evident in SDLN with both infrared and green light launched separately. When only infrared light was launched into the PPLN there was no manifestation of beam spreading along z-axis, nor of any type of scattering up to an average incident power of about 30 mW. We then launched only green light into the PPLN and, from an output power level of about 3 mW up to about 10 mW, a broad and transient scattering appeared (Fig.2a). The polarization of the scattered light was the same as for the input beam (along the z-axis). The total scattered power in the steady state, and the power of the main output beam as a function of the total output power from the sample, are represented in Fig.3. For a steady-state total output power of  $\sim 10$  mW the transient response of the scattering in Fig.2.a is shown in Fig.4.

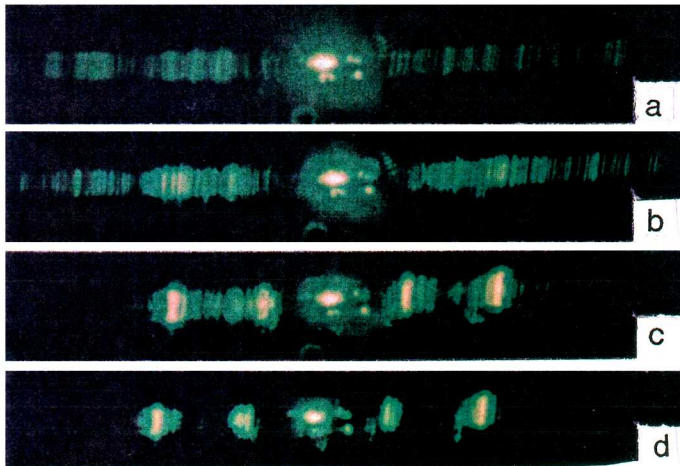


Fig.2 Far-field pattern of the output from the sample: (a) broad-transient scattering below threshold, (b),(c) intermediate situation at threshold, (d) steady-state 4 scattered orders above threshold.

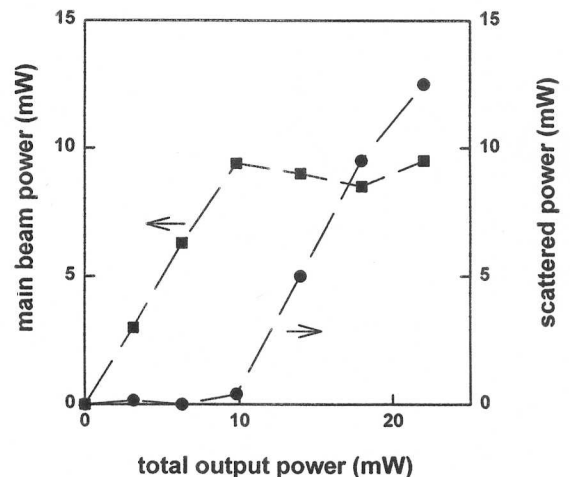


Fig.3 Steady-state total scattered power and main output power as functions of the total output power (measuring the main beam depletion).

Above 10 mW, four clear diffracted orders appeared (Fig.2d). The polarization of the scattering was still the same as that of the main beam and essentially all the excess pump power above 10 mW was scattered. This steady state scattering grew exponentially, typically

in 100 ms after the start of the illumination.

The power level of 10 mW is the threshold at which the broad and transient scattering transformed into a steady-state response with four clear diffracted orders. We observed this transformation and its reversal (Fig.2b and Fig.2c) at a fixed power of about 10 mW; this was probably due to slight power fluctuations of the laser source, taking the process just above and just below threshold.

These results were obtained at room temperature. Using a temperature-controlled oven we found that the threshold increased with the temperature. At about 28°C for a total output power of 22 mW, the steady-state scattering basically disappeared while this happened at about 50°C for the transient scattering. We point out that this is a remarkable change respect to SDLN where the photorefractive damage usually disappears at about 150°

Basically the same phenomena were observed in sample B , with one main difference: in sample A the external angles (with respect to the direction of propagation) relative to the first and second order were 12° and 26° while for sample B they were 9.5° and 20°. We estimate that these values correspond to a transverse refractive index grating in the crystal with a period of about 2.5 μm for sample A and 3.2 μm for sample B. The grating period is thus a constant fraction ~1/2.8 of the pitch of the domain inversion.

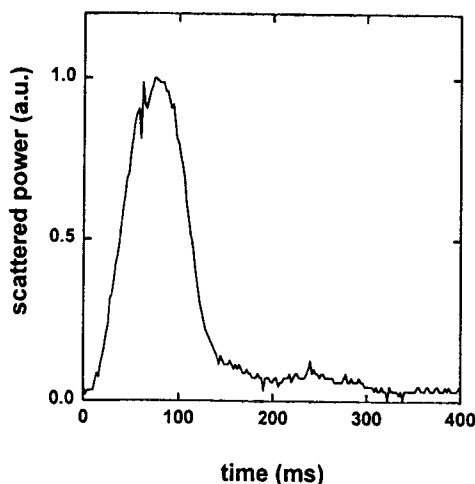


Fig.4 Transient response of the broad scattering. The growth and relaxation times scale inversely with the power (from curves at different powers).

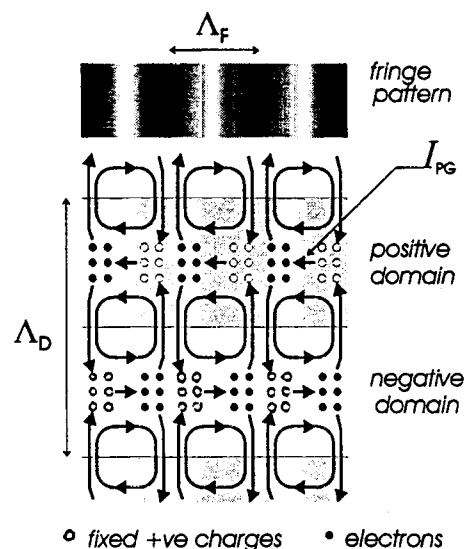


Fig.5 Current loops driven by the photo-galvanic effect for two-beam interference pattern. For five beam coupling the situation will be more complex.

When infrared and green light were used simultaneously, only two orders of scattering were observed for the infrared, coinciding spatially with the two 2nd order beams of the green (expected if both beams are scattered by the same refractive index grating).

We believe that the origin of this self-organised light-induced scattering is the photo-galvanic effect (see Figure 5), which causes a current to appear along the z direction in proportion to the 532 nm intensity [1]. This is consistent with the dependence on the wavelength (nothing occurred with only infrared light) and on the polarization state of the light. At lower power levels (3 to 10 mW), the photo-galvanic effect causes unpredictable spatial variations in space charge field and hence (via the Pockels effect) refractive index, which gives rise to the observed multi-directional transient scattering. This scattered light, acting as a noise source, seeds the growth of a refractive index grating which reinforces

scattering of the pump beam in certain preferred directions. We conjecture that the cycle of cause and effect is as follows. Interference between pump and noise beamlets gives rise to fringe patterns; photo-galvanic charge transfer across each fringe causes the refractive index to change at the intensity maxima; the refractive index grating thus formed diffracts light in specific directions, amplifying the noise beamlets in these directions. Self-reinforcing feedback is provided through reflection off the output, side and inner input faces of the sample. Above a certain pump power level this feedback is sufficient to cause strong nonlinear coupling of power into the four observed diffracted orders. We estimated that the first order and second order scattered beams reflect internally against the side faces at least three and six times before leaving the sample. The interaction length of the second order beam with the transverse refractive index grating is then about twice the interaction length of the first order. Probably this is why the second order spot appears brighter than the first order one.

There remains the question of why these directions are preferred, and why they scale with the inverse of the domain period. An answer lies in the fact that the direction of the photo-galvanic current  $I_{PG}$  changes from domain to domain; this will cause stable current loops to appear centred at the domain boundaries (Fig. 5). It is likely that these loops are stable only for a certain aspect ratio (which turns out to be when the ratio of domain period to fringe spacing is  $\sim 2.8$ ). The diffracted orders corresponding to fringes with this spacing experience the maximum gain and reach threshold first. Although the process bears some resemblance to subharmonic formation in two-beam coupling [9], the usual arguments based on two-beam coupling cannot be relied upon since the effect involves five beams and appears to be *local*.

In conclusion, a new kind of self-organised light-induced scattering typical of PPLN has been discovered. As the pump power is raised, random spatial instabilities appear and evolve, at a distinct nonlinear threshold power, into a stable pattern of diffracted beams. The pump beam itself is not distorted by these instabilities, which appear at quite large angles (light scattering at small angles, where the period of the interference pattern is wider than the period of the domain structure, is not effective in PPLN).

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