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Intensity Dependence of Thermal Nonlinear Optical Activity in Crystals

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Abstract. The thermal contribution to nonlinear optical activity in a BSO crystal was studied at 514.5 nm. The results show that the effect is only weakly dependent on laser intensity for constant beam power. By varying the laser spot size in the sample the electronic and thermal contributions to nonlinear optical activity may be separated.

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Natural optical activity (OA) in a crystal (rotation of the plane of polarization due to the spatial dispersion of the medium) depends on the temperature of the sample. An intense light beam can heat the crystal by absorption and so can change the angle of rotation [1]. This is one very simple mechanism of Nonlinear Optical Activity (NOA) and in gyrotropic crystals it should be taken into consideration along with the fast electronic NOA [2], especially in the case of strong absorption.

We report here, for the first time, that for TNOA with cw lasers, the angle of nonlinear rotation is independent of light beam intensity (for constant beam power) but depends on power.

1. Introduction and History

Thermal nonlinear rotation of the plane of polarization was the first mechanism of NOA experimentally studied in crystals. Vlasov and Zaitseva [1] carried out NOA experiments in coloured crystalline quartz with a pulsed ruby laser. Strong nonlinear optical activity and self-focussing was found in Bismuth Silicon Oxide (BSO) and Bismuth Germanium Oxide (BGO) by Bairamov et al. [3, 4]. This was qualitatively explained by a mechanism based on the resonance contribution of two-photon transitions involving real impurity states acting on intermediate levels. The fast electronic NOA was first isolated from the thermal background by Akhmanov et al. [5] in experiments on Lithium Iodate crystals. Pump-probe TNOA experiments were carried out cholesteric liquid crystals by Garibyan et al. [6]. It was observed that the destruction of a helix owing to the laser heating may be of a reversible nature. The information arising from the dynamics of the change in gyrotropy can be used for studying the rate of crystal-structure reconstruction in the presence of seeds.

2. Theory

The physical descriptions of TNOA with pulsed and cw lasers are different in principle. With short single light pulses ($\Delta t < S\varrho r^2/s$, where S is the specific heat of the crystal, ϱ is the density, s is the coefficient of thermal conductivity and r is the radius of the light beam in the sample) the energy absorbed from a pulse does not have time to leave the excited zone during the pulse. The temperature change depends on the energy density absorbed from the laser beam and thus on the laser beam intensity I. The nonlinear change in the specific constant of optical rotation, g(TNOA), is proportional to the temperature change and the coefficient of the temperature dependence of gyrotropy (dg/dT).

With very short light pulses ($\Delta t < r/v$, v is the speed of sound in the sample) the coefficient of the temperature dependence of gyrotropy should be taken at a constant volume, $(dg/dT)_v$, because the crystal has not time to expand during the light pulse. For longer pulses

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$(r/v < \Delta t < S\varrho r^2/s)$ the coefficient of the temperature dependence of nonlinear optical activity should be taken at a constant pressure $(dg/dT)_p$. In both cases we have:

$$g(\mathrm{TNOA}) \propto (dg/dT)_{v,p} I(z) \Delta t k/S,$$

where k is the absorption coefficient. One can see that the thermal nonlinear addition, g(TNOA), to the specific gyrotropy, g(OA), with pulsed lasers is proportional to light intensity I.

In stationary and quasi-steady state cases $(\Delta t < S\varrho r^2/s)$ the situation is different: the temperature of the excited volume depends on the balance between cooling and laser heating. To determine the temperature distribution in the sample, it is necessary to know how the generated heat is conducted away from the laser beam. For plane parallel samples two cooling mechanisms are important: heat conduction in the plane of the sample and heat conduction by surrounding air if any.

Hajto and Janossy [7] showed that in both cases the spot temperature is governed by the equation:

$$dT/dt = I[1-\exp(-kL)]/pSL - [T-T(0)]/\tau, \qquad (1)$$

where τ is an effective cooling time, different for various mechanisms of cooling, L is the sample thickness and the crystal temperature $T(0)$ is assumed to be fixed at the planar radius R. Under steady state conditions $(dT/dt = 0)$, and for low absorption, $(kL) < 1$, the temperature change is:

$$T - T(0) = Ik\tau/pS. \qquad (2)$$

If we can change the spot radius r of a Gaussian laser beam without changing the total laser beam power P we find (first order approximation) that the intensity I and the cooling time τ decrease and increase respectively as the square of the radius r. This means that the temperature of the sample in the centre of the laser spot does not depend on intensity for a fixed beam power. More detailed calculations show, that the cooling time, τ, is equal to $\tau'[1+2\ln(R/r)]$ when τ' is a function of r^2. Thus the temperature of the crystal and hence the angle of optical rotation θ(TNOA), for a crystal of thickness L, depends only logarithmically on the laser spot radius:

$$\theta(\mathrm{TNOA}) = L(dg/dT)[T-T(0)]$$
$$= [L(dg/dT)Pkp/pS][1+2\ln(R/r)]. \qquad (3)$$

In (3), $p = \tau'/r^2$ and is a constant depending on the thermal conductivity and the shape of the sample.

The weak intensity dependence means that results from cw TNOA experiments should preferably be presented in "angle.length^{-1}.power^{-1}" (deg mm^{-1} W^{-1}) units, but not in "angle.length^{-1}.intensity^{-1}" (deg mm W^{-1}) units which are more suited to pulsed NOA data. It should be noted that power-dependent thermal NOA effects can thus be detected in gyrotropic crystals under relatively low light intensities and so without strong temperature gradients and related stresses in the sample.

3. Experimental

Thermal nonlinear optical activity was studied in a planar BSO crystal of 1.6 mm thickness, grown by GEC Hirst Research Centre. BSO is a cubic crystal and is thus not naturally birefringent but it does exhibit strong isotropic optical activity. The light was incident on the crystal along the [110] direction.

The experimental arrangement is shown in Fig. 1. The sample was contained in an Oxford Instruments DN 704 cryostat which enabled the temperature to be adjusted in the 80–300 K range if necessary. A cw argon-ion laser, equipped with an acousto-optic mode-locker was used as the light source. High quality film polarisers were used to produce and analyse the state of polarisation. In these experiments, the light beam power was recorded after the analyser as a function of the angle between it and the polariser. Analyser orientation was controlled from a microcomputer by a stepper motor and varied typically by $\pm 1°$ near the crossed position. In this region the dependence of intensity on the angle between polariser and analyser is very close to a parabola. The bottom point of the parabola is the analyser orientation corresponding to the extinction of the light emerging from the sample.

A Bentham Model 218 lock-in amplifier also controlled by the microcomputer and a silicon photodiode with an Ulbricht-type photometric integrator were used for accurate intensity measurements. Typically 100–200 points, with 5–10 s averaging time for each point, were recorded near the bottom of the parabola. This was sufficient to fix the lowest point and hence the angle of optical activity in the sample by means of a real-time least-square polynomial approximation with an accuracy of better than 0.1°.

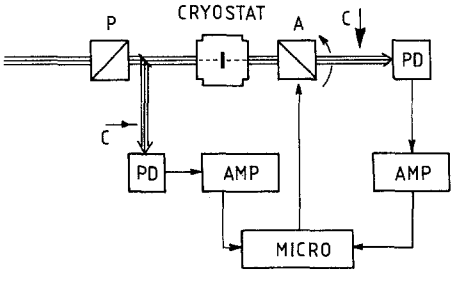

Fig. 1. Experimental arrangement for the study of nonlinear optical activity: P – polariser, A – analyser, C – chopper positions in the signal and reference beams, PD – photodiode, AMP – Lock-in amplifier, MICRO – microcomputer

4. Results and Discussions

TNOA was studied in detail for the 514 nm line of the argon-ion laser. The Gaussian beam was focussed into the sample by a 100 mm focal length lens and the cross-sectional area was varied by change of sample position with respect to this lens. In Fig. 2 the angle of optical activity as a function of the laser power for different beam cross-sections is shown. Curves A, B, C, and D correspond to spot diameters, $2r$, equal to 348 μm, 202 μm, 103 μm, and 27 μm respectively, i.e. the laser intensity increased by a factor of 167. Measurements of TNOA for laser powers greater than 200 mW was avoided because of the possibility of crystal damage. One can see that, as predicted by the phenomenological calculations above, the nonlinear rotation of the plane of polarization is proportional to the beam power but depends weakly on cross-sectional area i.e. light intensity. As discussed above, Eq. (3) shows that there is a logarithmic dependence of the TNOA parameter, θ/PL, on the beam spot radius in the sample. This is confirmed in Fig. 3 in which the TNOA parameter is shown plotted against $\ln(R/r)$ yielding a straight line.

The results presented in Figs. 2 and 3 constitute very strong evidence for the thermal nature of the nonlinear optical activity in BSO. Nevertheless nonlinear optical rotation in BSO was attributed to an electronic mechanism of NOA in earlier work [3]. We performed a very simple experiment to isolate the fast electronic contribution to the effect. As already noted, the argon-ion laser was equipped with an acousto-optic mode-locked which enabled the peak intensity to be changed by ~50 times without change in the average laser power by switching from cw to mode-locked operation. No variation in the optical activity greater than the accuracy of the polarimeter (0.1 deg) was detected on switching between the cw and mode-locked (138 MHz, 100 ps) regimes at a constant average power of 100 mW for a 27 μm laser beam spot diameter in the sample. Thus the "fast" electronic NOA in BSO is less than 10^{-6} deg cm W^{-1}.

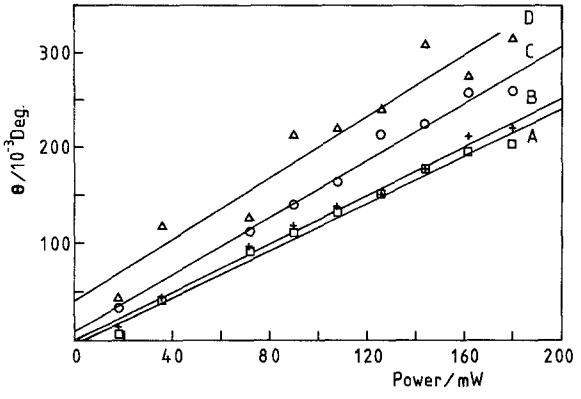

Fig. 2. The angle of rotation, θ, in BSO as a function of the argon-ion laser beam power at 514.5 nm for different beam cross-sectional areas at room temperature. Lines A, B, C, D refer to areas of 95000 μm^2, 32000 μm^2, 8300 μm^2, and 570 μm^2 respectively

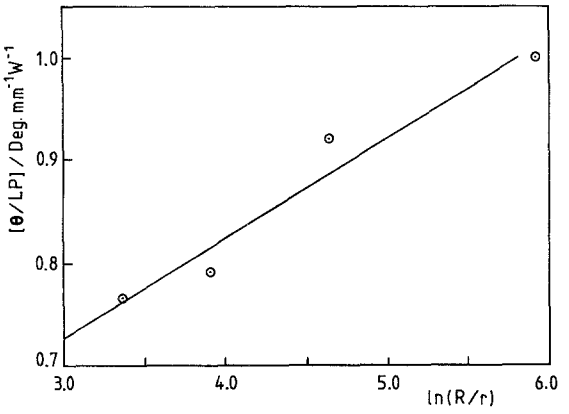

Fig. 3. TNOA, θ/PL, in BSO as a function of $\ln(R/r)$ where $R(=5$ mm) is the planar radius of the BSO sample and r is the spot radius of the argon-ion beam in the sample

More detailed results of TNOA in BSO will be presented in a subsequent paper.

5. Conclusions

Results have been presented concerning nonlinear optical activity caused by thermal self-action in crystals. By varying the laser spot size in the sample it has been demonstrated that the effect is only weakly intensity dependent. These results show that NOA experiments conducted with the aim of studying the fast electronic response due to the spatial dispersion of the nonlinearity in gyrotropic crystals must be performed with extreme accuracy, bearing in mind that the thermal contribution may be significant. The problem of separating the electronic and thermal elements under strong absorption conditions may be solved by observing the dependence of the effect on laser beam spot size.

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