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Photothermoelastic response of zincblende crystals to radiation from a THz-frequency quantum cascade laser

P. Dean,[1,*] A. H. Awang,[2] I. Kundu,[1] R. Alhathlool,[1] S. P. Khanna,[1] L. H. Li,[1] E. H. Linfield,[1] and A. G. Davies[1]

[1]School of Electronic and Electrical Engineering, University of Leeds, Leeds LS2 9JT, UK
[2]Universiti Teknologi MARA (UiTM), 40450 Shah Alam, Selangor Darul Ehsan, Malaysia
Email: *p.dean@leeds.ac.uk

***Abstract*—We investigate the photothermoelastic response of ZnTe and GaP crystals irradiated by THz-frequency radiation from a quantum cascade laser. We present a full theoretical description of this interaction that agrees well with the measured response.**

I. Introduction And Background

The use of optically-sampled crystals for the detection of terahertz (THz) frequency radiation has become widespread within applications such as THz time-domain spectroscopy. The most commonly adopted detection scheme exploits the linear electro-optic (EO) (or 'Pockels') effect in noncentrosymmetric crystals whereby the THz field induces a birefringence in the crystal (typically ZnTe) that can be probed optically. Electro-optic crystals have also been applied to incoherent and coherent sampling of THz fields generated using a quantum cascade laser (QCL) source. Interestingly, an incoherent interaction mechanism of thermal origin has recently been identified using a standard EO sampling arrangement with a ZnTe crystal and a THz QCL source [1].

In this paper we further investigate this interaction in ZnTe and GaP crystals illuminated by a QCL source emitting at 2.2 THz. Our results indicate a photothermoelastic origin of the interaction, whereby the stress distribution established through localised heating of the crystal induces a change in optical birefringence via the photoelastic response of the crystal. We have developed a comprehensive model of this previously unexplored mechanism, which shows good agreement with experimental data.

II. Experiment and Results

The photothermoelastic response of (110)-orientated ZnTe and GaP crystals with thicknesses L=1.9 mm and 1 mm, respectively, were investigated experimentally using a QCL source emitting at 2.2 THz. Radiation from the QCL was focused onto each crystal, and a 778 nm probe beam was employed to sample the induced birefringence optically using a balanced sampling arrangement [2]. Both the THz beam and the probe beam were linearly polarized parallel to the [-1,1,0] direction of the crystal, which in turn was oriented parallel to the polarisation axis of the Wollaston prism. The QCL device was operated in pulsed mode, emitting an average power ~845 μW. The pulse trains were electrically modulated in the frequency range 10 Hz–3 kHz, with lock-in detection of the photodiode response being employed.

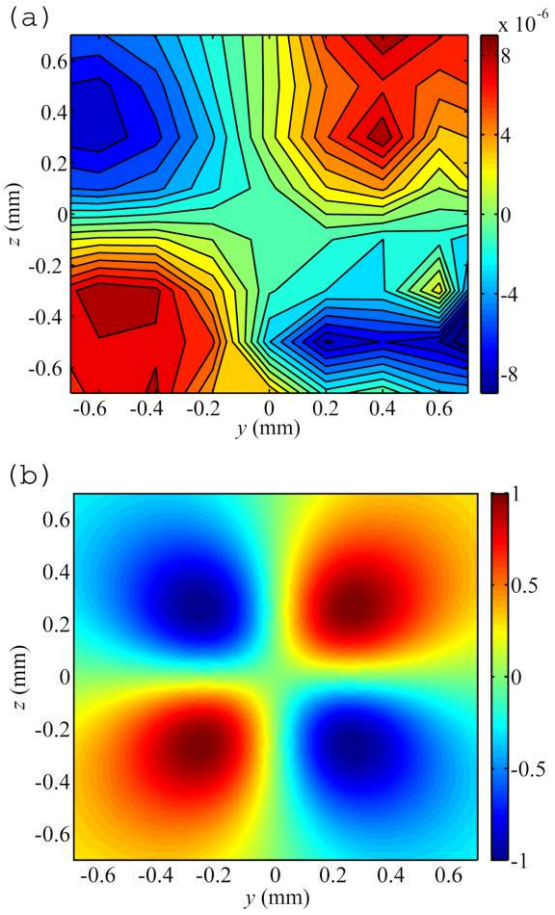

Fig. 1. (a) Contour plot showing the spatial variation of the photodiode signal measured across the y–z plane (the (110) plane) of the ZnTe crystal. The origin (0,0) corresponds to the centre of the THz beam. (b) Spatial variation of the (normalized) photodiode signal calculated using the photothermoelastic model for the same experimental conditions.

By translating the focusing lens in our apparatus, the probe beam could be scanned across the crystal surface in two dimensions. Using this approach the spatial variation of the measured response, relative to the position of the focussed THz beam, was investigated. Figure 1(a) shows a contour plot of the photodiode signal measured across an area of 1.4 mm ×

1.4 mm on the surface of the ZnTe crystal (defined as the y−z plane), for a laser modulation frequency of 40 Hz. As can be seen, the sign of the signal alternates in adjacent quadrants of the crystal surface and reaches a maximum magnitude at a critical radius along the ±45º diagonals. The intensity is also seen to vanish towards zero at the centre of the THz beam.

The absolute value of the phase delay between orthogonal components of the probe beam can be obtained from the photodiode response. Fig. 2 shows the optical phase delays measured as a function of laser modulation frequency, at the positions of maximum response (see Fig. 1), for the ZnTe and GaP crystals. As can be seen, the measured response decreases with modulation frequency ω and tends towards an inverse relationship at higher frequencies. A similar behavior has been observed previously [1] using a 3.2 THz QCL, and is symptomatic of a thermal response.

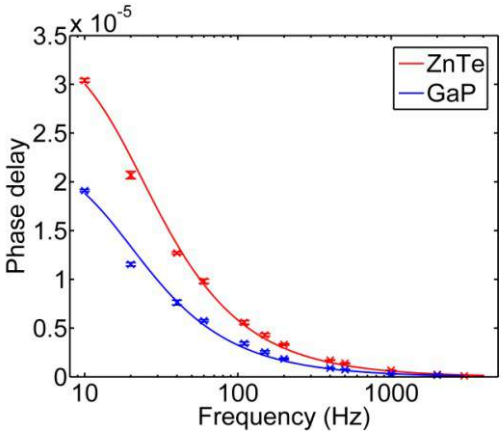

Fig. 2. Optical phase delay measured as a function of pulse modulation frequency for ZnTe and GaP crystals using a 2.2 THz QCL.

III. Theoretical Model and Discussion

We have developed a theoretical description of the measured response based on a photoelastic modulation of the optical birefringence arising from the stress distribution that is thermally-induced in the crystal upon localised THz irradiation. Our model predicts a modulation of the birefringence given by the relationship

$$\Delta n = \frac{-n^3}{2}\left[\frac{\left(\Delta B_2 - \Delta B_3\right)^2}{2\Delta B_4} + 2\Delta B_4\right]\sin\left(2\alpha\right), \qquad (1)$$

in which n is the refractive index and α is the angle between the [-1,1,0] direction of the crystal (the y−axis) and the major axis of the index ellipsoid induced through the photothermoelastic mechanism. The tensor components of the perturbation to the optical indicatrix, expressed in single-suffix (matrix) notation, are given by

$$\Delta B_i = p_{ij} s_{jk} \sigma_k, \qquad (2)$$

where p_{ij} are the components of the forth order photoelastic tensor and s_{jk} are the components of the fourth order elastic compliance tensor. The components of the second rank stress tensor σ_k are obtained by modeling the crystal as an isotropic thin disk subject to a radial temperature distribution determined by the Gaussian profile of the THz beam [3]. As such, Δn, ΔB_i and σ_k vary in three dimensions by virtue of attenuation of the THz power as the beam propagates through the crystal, as well as through the spatial distribution of the stress field in the y−z plane (the (110) plane of the crystal).

Figure 1(b) shows the spatial variation of the photodiode signal predicted using our model for the ZnTe crystal. The prediction is seen to agree well with the experimentally-determined spatial variation shown in Fig. 1(a). It should be noted that such a spatial variation is dependent upon the establishment of a radially-varying stress field in the (110) plane of the anisotropic crystal, and would not arise through a simple temperature-dependence of the refractive indices.

The measured response depends not only on the photoelastic properties of the crystal but also on the magnitude of the temperature modulation induced by the incident THz radiation. Our model also allows us to define a figure-of-merit for the response, at the position corresponding to the maximum signal, that depends on crystal properties including the elastic constant s_{44}, the photoelastic constant p_{44}, the volumetric heat capacity ρC, the thermal expansion coefficient α_{th}, the THz absorption coefficient α and Young's modulus Y:

$$FOM = \frac{\alpha_{th} Y n^3 p_{44} s_{44}\left(1 - e^{-\alpha L}\right)}{\rho C} \qquad (3)$$

For ZnTe and GaP we obtain values for the *FOM* of 6.9×10^{12} $\mathrm{J^{-1}m^3}$ and 3.0×10^{-12} $\mathrm{J^{-1}m^3}$ at 2.2 THz, respectively (see Fig. 1). By comparison, the measured phase delays for these crystals are $\delta = 6.7 \times 10^{-7}$ and 3.0×10^{-7} at $\omega = 1$ kHz, in good agreement with the ratio of figures of merit.

IV. Conclusion

We have investigated the photothermoelastic response of ZnTe and GaP crystals to QCL radiation at a frequency 2.2 THz, and developed a theoretical description of this mechanism that models the experimental data well.

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