Photoelastic Effect in Piezoelectric
Semiconductor : ZnO

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Photoelastic effect in piezoelectric semiconductor: ZnO

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The photoelastic tensor elements of the piezoelectric semiconductor ZnO were measured by the comparative acousto-optic diffraction method (reference material, fused quartz) at an acoustic frequency of 250 MHz and an optical wavelength of 0.633 μm. Two crystals with different conductivities, one is highly conductive and the other is highly resistive, were used as samples. The measured photoelastic tensor elements for piezoeactive strains are different between these two samples. The difference is explained by the electro-optic contribution to the photoelasticity induced by the electric field accompanying the piezoeactive acoustic wave in the highly resistive sample and its screening by free carriers in the highly conductive sample. The contribution of the rotation of the volume element to the photoelasticity has been also observed in photoelastic tensor elements for shear strains.

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I. INTRODUCTION

In piezoelectric crystals, the photoelastic effect consists of two parts. One is called the direct effect which expresses the direct contribution of acoustic strain [or displacement gradient] to the photoelasticity, and the other is called the indirect effect which expresses the macroscopic two-step contribution to the photoelasticity through piezoelectric and electrooptic effects. As was described in the paper by Nelson and Lax, the indirect effect had been ignored until Coquin pointed out the effect and its importance in ferroelectric lithium niobate in 1969, although the expression for the indirect photoelastic effect had been derived by Chapelle and Taurel in 1955. Recently, it was shown by White, Heidrich, and Lean that the indirect effect contributes dominantly to the acousto-optic interaction in a lithium niobate thin-film optical waveguide. So far, lithium niobate is the only material for which the direct and indirect photoelastic effects have been completely determined. Although it is expected that the photoelastic effect in the piezoelectric semiconductor should vary with the crystal conductivity because of the screening of the indirect contribution by free carriers, no experimental data have been found in literatures.5

In this paper we report the measurement of photoelastic tensor elements of ZnO crystal with different conductivities, one is highly conductive and the other is highly resistive. There are three reasons why ZnO was used: (i) ZnO is one of the most highly piezoelectric materials in piezoelectric semiconductors. (ii) A ZnO crystal with weak photoconductivity can be obtained. (iii) Most of the photoelastic tensor elements of ZnO besides ω4 are not known, therefore the experimental data are necessary in the field of acousto-optics, especially in ZnO thin-film acousto-optic devices. Measurements were performed at an acoustic frequency of 250 MHz and an optical wavelength of 0.633 μm. The measured photoelastic tensor elements for piezoeactive strains are different between these two samples, although the other elements for piezoeactive strains agreed within the experimental error. The difference is explained by the photoelasticity with and without the indirect effect depending on the crystal conductivity.

The contribution of the rotation of the volume element to the photoelasticity which was pointed out theoretically by Nelson and Lax and observed experimentally in rutile by Nelson and Lazay is also observed in the photoelastic tensor elements for shear strain.

II. SAMPLES AND EXPERIMENTAL PROCEDURE

The ZnO crystals used in the experiment were grown hydrothermally by Sakagami and Wada. An as-grown crystal with a conductivity of 2.67 (Ω cm)−1 was used as a highly conductive sample. To obtain a highly resistive sample, lithium was diffused into one of the as-grown crystals at 900°C for 10 h in an oxygen gas flow. The conductivity after the diffusion of lithium was 2.5×10−11 (Ω cm)−1 in the dark. The conductivity increased to 5×10−15 (Ω cm)−1 due to photoconductivity when the sample was irradiated uniformly by the He-Ne laser with output power of 5 mW. From this increase of conductivity the local conductivity, when the sample was irradiated locally by the laser to measure the photoelastic tensor element, was estimated to be less than 10−8 (Ω cm)−1. This sample was used as a highly resistive sample. The samples are about 5 mm cube with one edge parallel to the c axis.

The dispersion of refractive indices was measured by Sakagami and Wada using the minimum deviation method. The indices at 0.633 μm were n1 = 1.9957 and n2 = 1.9795 for as-grown crystals and n1 = 2.0300 and n2 = 2.0130 for lithium-diffused crystals.

The experimental arrangement for measuring the photoelastic tensor elements is shown in Fig. 1. The principle of the measurement has been described by Dixon and Cohen. This arrangement is essentially the same as that proposed by them, but sound cell 2 and

![FIG. 1. Experimental arrangement for measuring photoelastic tensor elements.](image-url)
TABLE I. Measured photoelastic tensor elements of ZnO (optical wavelength is 0.633 μm).

<table>
<thead>
<tr>
<th>Mode</th>
<th>Acoustic wave Propagation</th>
<th>Displacement</th>
<th>Piezoeactive Propagation</th>
<th>Optical wave Polarization</th>
<th>$\rho_{1111}$</th>
<th>Photelastic tensor elements</th>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Highly resistive sample ($\sigma$ is less than $10^{-5}$Ω/cm)</td>
</tr>
<tr>
<td>$L$</td>
<td>$X$</td>
<td>$X$</td>
<td>No</td>
<td>$\approx Y^a$</td>
<td>$X$</td>
<td>$\rho_{1111}$</td>
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<tr>
<td>$L$</td>
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<tr>
<td>$L$</td>
<td>$Z$</td>
<td>$Z$</td>
<td>Yes</td>
<td>$\approx Y^a$</td>
<td>$Z$</td>
<td>$\rho_{1111}$</td>
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<tr>
<td>$S$</td>
<td>$X$</td>
<td>$Z$</td>
<td>Yes</td>
<td>$\theta_\parallel$ to $Y$</td>
<td>$Z$ or</td>
<td>$\rho_{1111}$</td>
</tr>
<tr>
<td>$S$</td>
<td>$Z$</td>
<td>$X$</td>
<td>No</td>
<td>$\theta_\parallel$ to $Z$</td>
<td>$X$ or</td>
<td>$\rho_{1111}$</td>
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</table>

$^a$are used because the Bragg angle for longitudinal wave is less than 1$^\circ$.

$^b$Bragg angle ($\theta_\parallel$) for ordinary wave $\theta_\parallel$ and extraordinary wave $\theta_\parallel$ are 15.44$^\circ$ and 17.05$^\circ$ for the highly conductive sample and 17.15$^\circ$ and 18.68$^\circ$ for the highly resistive sample, respectively.

$^c$Then $\theta_\parallel$ is 13.95$^\circ$ and $\theta_\parallel$ is 15.55$^\circ$ for the highly conductive sample, and $\theta_\parallel$ is 15.03$^\circ$ and $\theta_\parallel$ is 16.61$^\circ$ for the highly resistive sample.

To deduce the values of photoelastic tensor elements from the experimental data, the elastic, piezoelectric and dielectric constants of ZnO given by Smith$^{42}$ were used as well as the refractive indices described before.

III. RESULTS AND DISCUSSION

Measured values of the independent photoelastic tensor elements are shown in Table 1. As shown in the table, the values of photoelastic tensor elements for piezoelectric strains, i.e., $\rho_{1111}$, $\rho_{1112}$, $\rho_{1113}$, and $\rho_{1133}$ agree well between the two samples. On the contrary, the values of the photoelastic tensor elements for piezoelectric strains, i.e., $\rho_{1112}$, $\rho_{1133}$, and $\rho_{1134}$ are considerably different between two samples. To explain this difference, let us consider the photoelastic effect in piezoelectric semiconductors.

The photoelastic effect in piezoelectric (therefore electro-optic) material is described as follows$^{18}$ (omitting, for a moment, the contribution of the rotation of the volume element described later):

$$\Delta(1/\kappa) = \rho_{1111}S_{11} + \rho_{1111}S_{11} + \rho_{1112}S_{12} + \rho_{1113}S_{13},$$  \hspace{1cm} (1)

where $\kappa$ is dielectric constant, $\rho_{1111}$ is the photoelastic (strain-optic) tensor measured at constant electric field, $S$ is acoustic strain, $\rho_{1112}$ is the electro-optic tensor measured at constant strain, and $E$ is the electric field induced by the piezoelectricity of the crystal. The first and second terms of Eq. (1) express direct and indirect photoelastic effects, respectively. The indirect photoelastic effect is a two-step contribution to the photoelasticity through piezoelectric and electro-optic effects. On the other hand, the electric field which accompanies the acoustic wave within a piezoelectric semiconductor can be described as follows$^{19}$:

$$E_x = \frac{a_0 a_e e_p e_{11} S_{11}}{a_0 a_e a_e} \frac{1}{1+j\omega \omega_p} \frac{1}{1+j\omega \omega_p},$$  \hspace{1cm} (2)

where $a$ is the component of the unit vector $a$ of the acoustic wave, $e^p$ is the low-frequency dielectric tensor measured at constant strain, $\omega_0 = \sigma / e^p$, $\sigma$ is conductivity, $\omega_p = e^p q / \mu k T$, $q$ is electronic charge, $v$ is velocity of the acoustic wave, $\mu$ is carrier mobility, $k$ is the Boltz-
mann constant, and $T$ is absolute temperature) is the diffusion frequency. From Eqs. (1) and (2), the photoelastic effect in the piezoelectric semiconductor is written as follows:

$$\Delta \left( \frac{1}{\omega_0} \right) = \left( p_{11}^{\mu} \frac{T_{11}^d}{\rho_0} + \frac{1}{1 + j(\omega_0/\omega + \omega/\omega_0)} \right) S_{12}$$

$$= p_{11}^{\mu} + p_{11}^{\rho} S_{12}. \quad (3)$$

The indirect photoelastic effect in the piezoelectric semiconductor becomes a function of crystal conductivity. The variation of the indirect contribution with crystal conductivity is illustrated in Fig. 2. As shown in the figure, the indirect contribution vanishes at the highly conductive limit because the electric field induced by the piezoelectricity is completely screened by free carriers. Therefore, the photoelectric tensor measured at the highly conductive limit is $p_{11}^{\mu}$. On the contrary, the indirect effect contributes completely to the photoconductivity at the highly resistive limit. The photoelastic tensor measured at the highly resistive limit is referred to as $p_{11}^{\rho}$ because the electric displacement within the crystal is constant. $p_{11}^{\rho}$ is written as

$$p_{11}^{\rho} = p_{11}^{\mu} - \frac{T_{11}^d}{\rho_0} \frac{1}{1 + j(\omega_0/\omega + \omega/\omega_0)}. \quad (4)$$

The measure of the indirect contribution to the photoconductivity is a ratio of electric field $E$ to that when there are no free carriers $E_0$:

$$\frac{E}{E_0} = \frac{1 + j\omega_0/\omega + \omega/\omega_0}{1 + \frac{1}{1 + j(\omega_0/\omega + \omega/\omega_0)}}. \quad (5)$$

The value of $\omega_0/\omega$ for the highly conductive sample is 220 and that of $\omega/\omega_0$ is less than 0.1. Then, $E/E_0$ is about 0.005. It means that the electric field induced by the piezoelectricity is almost completely screened by free carriers. Therefore, the photoelastic tensor elements measured for the highly conductive sample should be $p_{11}^{\mu}$. On the contrary, the value of $\omega_0/\omega$ for the highly resistive sample is less than $10^{-6}$. Then, $E/E_0$ is equal to 1.0. Therefore, the photoelastic tensor elements measured for the highly resistive sample should be $p_{11}^{\rho}$. The indirect contribution at the highly resistive limit can be calculated from the electro-optic data given by Turner$^{18}$ ($r_{33}^{33} = 2.6 \times 10^{-12}$ m/V, $r_{11}^{33} = -1.4 \times 10^{-11}$ m/V) and from the piezoelectric dielectric data given by Smith$^{15}$ ($\epsilon_{33} = 1.321$ C/m$^2$, $\epsilon_3 = 9.03 \times 10^{-11}$ F/m) as follows:

$$p_{11}^{33} = -v_{33}^{33} \epsilon_3 / \epsilon_3 = +0.0205,$$

$$p_{33}^{33} = -v_{33}^{33} \epsilon_3 / \epsilon_3 = -0.038.$$  

The signs of $p_{1133}$ and $p_{3333}$ in Table I are determined from the signs of the indirect contribution calculated above. The measured indirect contributions are $p_{1133}^{\mu} = +0.021$ and $p_{3333}^{\mu} = -0.281$. Although the measured and calculated values of $p_{1133}^{\mu}$ cannot be compared because $v_{33}^{33}$ is not known, measured and calculated values of $p_{1133}$ and $p_{3333}$ can be compared, and it can be said that the difference of the measured photoelastic tensor elements for piezoelectric carrier conductivity and the highly resistive samples is the difference between $p_{11}^{\mu}$ and $p_{11}^{\rho}$.  

There is a possibility that the carrier-density wave induced by the piezoelectricity in the piezoelectric semiconductor contributes to the diffraction of light because the dielectric constant is perturbed by the carrier-density wave. In one-dimensional description, the perturbation of the free-carrier density induced by an acoustic wave in the piezoelectric semiconductor is written as follows$^{10}$:

$$\frac{n_s}{n_0} = \frac{1}{1 + j(\omega_0/\omega + \omega/\omega_0)} S,$$

where $n_s$ is space-charge density and $n_0$ is free-carrier density at equilibrium. On the other hand, the dielectric constant at a wavelength far from the band edge is related to the free-carrier density $n$ as follows$^{11}$:

$$\kappa(n) = \kappa(0) - n q^2 / \kappa_m \omega_0^2,$$

where $\kappa(0)$ is dielectric constant without free carriers, $q$ is electronic charge, $\kappa_0$ is the permittivity of a vacuum, $m_e$ is free-electron mass, and $\omega_0$ is the optical frequency. We neglect here the imaginary part of the dielectric constant, i.e., the term for free-carrier absorption, because it does not contribute to the diffraction of light in the Bragg region. From Eqs. (6) and (7), the perturbation of $1/\kappa$ can be written as

$$\Delta(1/\kappa) = \frac{1}{1 + j(\omega_0/\omega + \omega/\omega_0)} \left[ \frac{q^2 n_s}{\kappa_0 m_e \omega_0^2} \right] = p^{\text{free}} S.$$

$p^{\text{free}}$ is the contribution of the carrier-density wave to the photoelastic effect. Numerical calculation for the sample used in the experiment revealed that $p^{\text{free}}$ is an order of magnitude less than $p^{\text{ind}}$. Therefore, $p^{\text{free}}$ does not contribute to the difference of the photoelastic tensor elements measured for the highly conductive and highly resistive samples. The free-carrier contribution will become significant at a much longer wavelength of light.

Next, let us consider the difference between $p_{1133}$ and $p_{1133}^{\mu}$ measured for the highly conductive sample. These tensor elements had been described by the same abbreviated notation ($p_{11}^{\mu}$) until the contribution of the rotation of the volume element to the photoelasticity was pointed out by Nelson and Lax.$^1$ From their theory, the contribution of the rotation of the volume element is
written as

\[ P_{\langle i j k \rangle 123} = \frac{1}{2} \left[ (\kappa^{-1})_{ij} \delta_{kl} + (\kappa^{-1})_{ij} \delta_{lk} - (\kappa^{-1})_{ik} \delta_{lj} - (\kappa^{-1})_{ik} \delta_{lj} \right]. \]

Using the refractive indices measured for the highly conductive sample \( n_\parallel = 1.9795 \) and \( n_\perp = 1.9957 \), \( p_{1313131} \) and \( p_{1313131} \) are calculated as \( p_{1313131} = -0.0021 \) and \( p_{1313131} = +0.0021 \). Therefore, the difference between \( p_{1313} \) and \( p_{1313} \) becomes \( -0.0002 \). The signs of the measured photoelastic tensor elements \( p_{1313} \) and \( p_{1313} \) are determined from the sign of this contribution of the rotation of the volume element. The difference between measured \( p_{1313} \) and \( p_{1313} \) is \( -0.005 \), and it is in good agreement with the calculated value within the experimental error. For the highly resistive sample the measured values of \( p_{1313} \) and \( p_{1313} \) are the same. It means that the contribution of the rotation of the volume element and the contribution of the indirect photoelastic effect cancelled out each other.

IV. CONCLUSION

The direct and indirect photoelastic effects as well as the contribution of the rotation of the volume element to the photoelasticity in the piezoelectric semiconductor ZnO have been completely determined by measuring the photoelastic tensor element for both of the highly conductive and highly resistive samples, respectively. It was clarified that the values of the photoelastic tensor elements for piezoelectric strains vary between \( \rho^p \) and \( \rho^d \), depending on the crystal conductivity because of the screening of the two-step contribution to the photoelasticity through the piezoelectric and the electro-optic effects by free carriers.

ACKNOWLEDGMENTS

The authors are very grateful to N. Sakagami and M. Wada for supplying ZnO crystals and the experimental data of refractive indices of ZnO.

5) Preliminary experiment on the variation of the stress-optic constant of CdS with crystal conductivity has been reported by T. Sueda and T. Ogawa in the Spring Meeting of the Related Societies of Applied Physics in Japan, 28-G-11, 1973 (unpublished).
12) N. Sakagami and M. Wada (private communication).
18) E. H. Turner (unpublished), but data are inserted in Ref. 16, p. 463.
19) The contribution of the indirect effect in the ZnO crystal is less than 20% at most. Therefore, the theoretical calculations for the diffraction efficiency in ZnO thin-film acousto-optic devices (Refs. 8 and 9) in which the contribution of indirect effect was ignored are still valid approximation compared with the case of a lithium-niobate thin-film acousto-optic device (Ref. 4) in which the contribution of indirect effect is an order of magnitude larger than that of direct effect.