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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 $0.633 \mu\text{m}$. 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 piezoactive 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 piezoactive 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,¹ the indirect effect had been ignored until Coquin² 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 Chappelle and Taurel³ in 1955. Recently, it was shown by White, Heidrich, and Lean⁴ 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 p_{66} ⁶ are not known, therefore the experimental data are necessary in the field of acousto-optics, especially in ZnO thin-film acousto-optic devices.⁷⁻⁹ Measurements were performed at an acoustic frequency of 250 MHz and an optical wavelength of $0.633 \mu\text{m}$. The measured photoelastic tensor elements for piezoactive strains are different between these two samples, although the other elements for piezononactive 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 Lax¹ and observed experimentally in rutile by Nelson and Lazay¹⁰ 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.¹¹ An as-grown crystal with a conductivity of $2.67 (\Omega \text{cm})^{-1}$ 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 $2.5 \times 10^{-11} (\Omega \text{cm})^{-1}$ in the dark. The conductivity increased to $5 \times 10^{-10} (\Omega \text{cm})^{-1}$ 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 $10^{-8} (\Omega \text{cm})^{-1}$. 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 Wada¹² using the minimum deviation method. The indices at $0.633 \mu\text{m}$ were $n_e = 1.9957$ and $n_o = 1.9795$ for as-grown crystals and $n_e = 2.0300$ and $n_o = 2.0130$ 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.¹³ This arrangement is essentially the same as that proposed by them, but sound cell 2 and

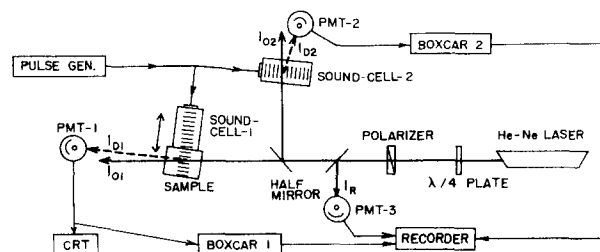


FIG. 1. Experimental arrangement for measuring photoelastic tensor elements.

TABLE I. Measured photoelastic tensor elements of ZnO (optical wavelength is 0.633 μm).

Mode	Acoustic wave			Optical wave		$p_{ij,kl}$	Photoelastic tensor elements	
	Propagation	Displacement	Piezoactive	Propagation	Polarization		Highly conductive sample ($\sigma = 2.67 \text{ } \Omega/\text{cm}$)	Highly resistive sample (σ is less than $10^{-8} \text{ } \Omega/\text{cm}$)
L	X	X	No	$\approx Y^a$	$\approx X$	$ p_{1111} $	0.222	0.221
L	X	X	No	$\approx Z$	Y	$ p_{1122} $	0.099	0.099
L	X	X	No	$\approx Y$	Z	$ p_{3311} $	0.088	0.089
L	Z	Z	Yes	$\approx Y$	X	p_{1133}	-0.111	-0.090
L	Z	Z	Yes	$\approx Y$	$\approx Z$	p_{3333}	-0.235	-0.263
S	X	Z	Yes	θ_B^b to Y	Z or θ_B to X	p_{1331}	-0.056	-0.061
S	Z	X	No	θ_B^c to Y	X or θ_B to Z	p_{1313}	-0.061	-0.061

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

^bBragg angle (θ_B) for ordinary wave θ_o and extraordinary wave θ_e are 15.44° and 17.05° for the highly conductive sample

and 17.15° and 18.66° for the highly resistive sample, respectively.

^c $\theta_o = 13.95^\circ$ and $\theta_e = 15.55^\circ$ for the highly conductive sample, and $\theta_o = 15.03^\circ$ and $\theta_e = 16.61^\circ$ for the highly resistive sample.

photomultiplier tubes, PMT-2 and PMT-3, are added to correct the fluctuations of the output powers of the rf-pulse generator and He-Ne laser. Fused quartz was used as a reference material. ZnO thin-film transducers for exciting longitudinal and shear waves were formed on two fused-quartz cubes, respectively, by the dc sputtering technique.¹⁴ These two fused-quartz cubes with ZnO thin-film transducers were used as sound cell 1. The ZnO thin-film transducer for exciting longitudinal waves was also formed on the CdS crystal, and it was used as sound cell 2. CdS was used because of its high figure of merit. The ZnO sample to be measured is bonded to sound cell 1. Acoustic waves with a frequency of 250 MHz are generated into sound cell 1 and sound cell 2 in parallel. The He-Ne laser beam is separated into three beams by half-mirrors as shown in the figure. The intensity of the beam diffracted in sound cell 2 (I_{D2}) and the intensity of the reference beam (I_R) are always detected by PMT-2 and PMT-3. The intensity of the beam diffracted in sound cell 1 or in the ZnO sample (I_{D1}) is detected by PMT-1 and it is normalized by I_{D2} . To measure the transmission coefficient of the ZnO sample or sound cell 1, the transmitted beam intensity (I_{01}) when the acoustic wave is absent is detected by PMT-1 and it is normalized by I_R . Thus, the fluctuations of the output powers of the He-Ne laser and rf-pulse generator can be completely cancelled out. To allow the diffraction efficiencies to remain linear with acoustic power, diffraction efficiencies I_{D1}/I_{01} and I_{D2}/I_{02} are kept less than 1%. Care must be taken when the photoelastic tensor elements which induce polarization rotation of light from ordinary waves to extraordinary waves or vice versa are measured, because the transmission coefficients at the crystal surface for ordinary and extraordinary waves are not equal, especially when the light beam passes through the crystal surface obliquely. This difference of transmission can be cancelled out if two measurements for polarization rotation from ordinary to extraordinary waves and vice versa are performed, and then the geometrical mean of the deduced apparent photoelastic constants is taken. Over-all accuracy of this experimental arrangement is tested by using a fused-quartz cube as a sample. The observed experimental error was less than 3%.

To deduce the values of photoelastic tensor elements from the experimental data, the elastic, piezoelectric and dielectric constants of ZnO given by Smith¹⁵ 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 I. As shown in the table, the values of photoelastic tensor elements for piezoelectric strains, i. e., p_{1111} , p_{1122} , p_{3311} , and p_{1313} agree well between the two samples. On the contrary, the values of the photoelastic tensor elements for piezoactive strains, i. e., p_{1133} , p_{3333} , and p_{1331} 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 follows¹⁶ (omitting, for a moment, the contribution of the rotation of the volume element described later):

$$\Delta(1/\kappa)_{ij} = p_{ij,kl}^E S_{kl} + r_{ij,k}^S E_k, \quad (1)$$

where κ is dielectric constant, p^E is the photoelastic (strain-optic) tensor measured at constant electric field, S is acoustic strain, r^S 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 follows¹⁷:

$$E_r = -\frac{a_r a_s e_{s,kl} S_{kl}}{a_p \epsilon_p a_q} \frac{1 + j(\omega/\omega_D)}{1 + j(\omega_c/\omega + \omega/\omega_D)}, \quad (2)$$

where a is the component of the unit wave vector \mathbf{a} of the acoustic wave, ϵ^S is the low-frequency dielectric tensor measured at constant strain, $\omega_c (= \sigma/\epsilon^S, \sigma$ is conductivity) is the dielectric relaxation frequency, $\omega_D (= qv^2/\mu kT, q$ is electronic charge, v is velocity of the acoustic wave, μ is carrier mobility, k is the Boltz-

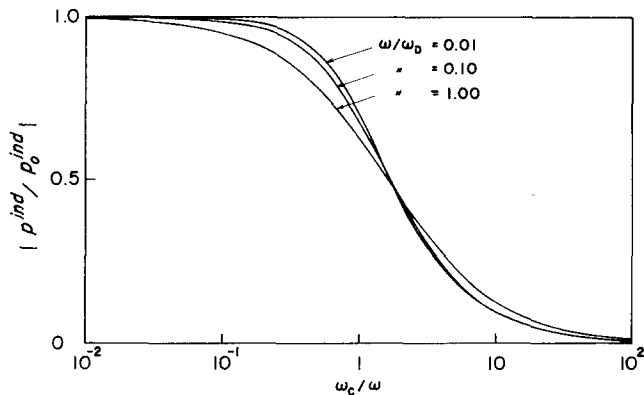


FIG. 2. Variation of indirect photoelastic effect with crystal conductivity. p_0^{ind} expresses the indirect photoelastic effect when crystal conductivity is zero.

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:

$$\Delta\left(\frac{1}{\kappa}\right)_{ij} = \left(p_{ij,kl}^E - \frac{r_{ij}^S r_{kl}^S a_r a_s e_{s,kl}}{a_p \epsilon_{pq}^S a_q} \frac{1 + j\omega/\omega_D}{1 + j(\omega_c/\omega + \omega/\omega_D)} \right) S_{kl} \equiv (p_{ij,kl}^E + p_{ij,kl}^{\text{ind}}) S_{kl}. \quad (3)$$

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 photoelastic tensor measured at the highly conductive limit is $p_{ij,kl}^E$. On the contrary, the indirect effect contributes completely to the photoelasticity at the highly resistive limit. The photoelastic tensor measured at the highly resistive limit is referred to as $p_{ij,kl}^D$ because the electric displacement within the crystal is constant. $p_{ij,kl}^D$ is written as

$$p_{ij,kl}^D = p_{ij,kl}^E - \frac{r_{ij}^S r_{kl}^S a_r a_s e_{s,kl}}{a_p \epsilon_{pq}^S a_q}. \quad (4)$$

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

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

The value of ω_c/ω for the highly conductive sample is 220 and that of ω/ω_D 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 $p_{ij,kl}^E$. On the contrary, the value of ω_c/ω 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 $p_{ij,kl}^D$. The indirect contribution at the highly resistive limit can be calculated from the electro-optic data given by Turner¹⁸ ($r_{333}^S = +2.6 \times 10^{-12}$ m/V, $r_{113}^S = -1.4$

$\times 10^{-12}$ m/V and from the piezoelectric dielectric data given by Smith¹⁵ ($e_{333} = +1.321$ C/m², $\epsilon_{33}^S = 9.03 \times 10^{-11}$ F/m) as follows:

$$p_{1133}^{\text{ind}} = -r_{113}^S e_{333} / \epsilon_{33}^S = +0.0205,$$

$$p_{3333}^{\text{ind}} = -r_{333}^S e_{333} / \epsilon_{33}^S = -0.038.$$

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 $p_{1133}^{\text{ind}} = +0.021$ and $p_{3333}^{\text{ind}} = -0.028$. Although the measured and calculated values of p_{1331}^{ind} cannot be compared because r_{311}^S is not known, measured and calculated values of p_{1133}^{ind} and p_{3333}^{ind} can be compared, and it can be said that the difference of the measured photoelastic tensor elements for piezoactive strains between the highly conductive and the highly resistive samples is the difference between $p_{ij,kl}^E$ and $p_{ij,kl}^D$.¹⁹

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.^{20,21} In a one-dimensional description, the perturbation of the free-carrier density induced by an acoustic wave in the piezoelectric semiconductor is written as follows¹⁷:

$$\frac{n_s}{n_0} = \frac{e\mu}{v\epsilon^S} \frac{1}{1 + j(\omega_c/\omega + \omega/\omega_D)} S, \quad (6)$$

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 follows²²:

$$\kappa(n) = \kappa(0) - q^2 n / \kappa_0 m_c \omega_0^2, \quad (7)$$

where $\kappa(0)$ is dielectric constant without free carriers, q is electronic charge, κ_0 is the permittivity of a vacuum, m_c is free-electron mass, and ω_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

$$\Delta\left(\frac{1}{\kappa}\right) = -\frac{e\mu}{v\epsilon^S} \frac{1}{\kappa_0 \kappa^2} \frac{q^2 n_0}{m_c \omega_0^2} \frac{1}{1 + j(\omega_c/\omega + \omega/\omega_D)} S \equiv p^{\text{fc}} S. \quad (8)$$

p^{fc} is the contribution of the carrier-density wave to the photoelastic effect. Numerical calculation for the sample used in the experiment revealed that p^{fc} is an order of magnitude less than p^{ind} . Therefore, p^{fc} 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_{1331} and p_{1133} measured for the highly conductive sample. These tensor elements had been described by the same abbreviated notation (p_{44}) 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

$$P_{(ij)[kl]} = \frac{1}{2}[(\kappa^{-1})_{il}\delta_{kj} + (\kappa^{-1})_{lj}\delta_{ik} - (\kappa^{-1})_{ik}\delta_{lj} - (\kappa^{-1})_{kj}\delta_{il}]. \quad (9)$$

Using the refractive indices measured for the highly conductive sample ($n_o = 1.9795$ and $n_e = 1.9957$), $p_{(13)[13]}$ and $p_{(13)[31]}$ are calculated as $p_{(13)[13]} = -0.0021$ and $p_{(13)[31]} = +0.0021$. Therefore, the difference between p_{1313} and p_{1331} becomes -0.0042 . The signs of the measured photoelastic tensor elements p_{1313} and p_{1331} are determined from the sign of this contribution of the rotation of the volume element. The difference between measured p_{1313} and p_{1331} 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_{1331} 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 piezoactive strains vary between p^E and p^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.

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