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Piezobirefringence analysis in an opaque region

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We report a new method to analyze the piezobirefringence coefficient in an opaque region in which the stress-induced changes in both the real ($\Delta\epsilon_1$) and imaginary part ($\Delta\epsilon_2$) of the dielectric constant are properly taken into account. New coefficients, which determine the fractional contributions of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ to the piezobirefringence coefficient, have been derived from an analytical point of view. The experimental data on Si and ZnSe reported recently have been analyzed by using the present result. Good agreement between the experiment and calculation has been found. The present method provides a guiding principle for analyzing the piezobirefringence coefficient in an opaque region.

I. INTRODUCTION

We recently reported the spectral dependence of the photoelastic constants $p_{11}-p_{12}$ and p_{44} in various semiconductors in the region below the lowest direct gap (E_0 gap) determined from the Brillouin-scattering measurements by introducing the intrinsic piezobirefringence analysis.[1-3] We have also shown that the dispersion of the photoelastic constants depends strongly on a lifetime broadening of the electronic states in solids. In the analysis, however, we have disregarded the contribution from the imaginary part of the dielectric constant to the photoelastic constant assuming that its contribution is negligibly small in the photon-energy region of transparency. Recently, Chandrasehkar, Grimsditch, and Cardona[4] developed a new method to measure stress-induced birefringence in an opaque region which employs the Raman-scattering technique as a probe. Using this new method, they have measured the magnitudes of the piezo-optical (photoelastic) constants in Si (0.5–3.38 eV)[5] and GaP (1.0–2.6 eV)[6] above the fundamental absorption edges. In their analysis, the contribution from the imaginary part of the dielectric constant has also been neglected, although they have pointed out that its contribution should be taken into account in order to discuss the spectral dependence of the piezo-optical constants in the region above 3.0 eV in Si.

In this paper, we report a new method to analyze the piezobirefringence effect in the opaque region of solids, where the stress-induced changes in both the real and imaginary parts of the dielectric constant are properly taken into account by introducing new fractional coefficients. From the present analysis, we again find that a close relation exists between the piezobirefringence (or Brillouin scattering) and modulation spectroscopy,[1,7] where the fractional coefficients in the former are very analogous in their behavior to the Seraphin coefficients in the latter. We will apply the present result to the analysis of the experimental data on Si (Ref. 5) and ZnSe (Ref. 1).

II. MODEL

When a uniaxial stress is applied to a crystal having diamond or zinc-blende structure the crystal becomes birefringent. The piezobirefringence is usually measured with linearly polarized light which propagates along a direction perpendicular to that of the applied stress.[8] Two linearly polarized waves with amplitudes $E_\parallel$ (parallel to the stress direction) and $E_\perp$ (perpendicular to the stress direction) will propagate through the crystal. The electric fields of these waves, as functions of the position x in the crystal, can be written as

$$E_\parallel(x)=E_0\exp[i(2\pi n_\parallel x/\lambda-\omega t)]\exp(-2\pi k_\parallel x/\lambda)\,, \tag{1a}$$

$$E_\perp(x)=E_0\exp[i(2\pi n_\perp x/\lambda-\omega t)]\exp(-2\pi k_\perp x/\lambda)\,, \tag{1b}$$

where E_0 is the amplitude of the incident wave, and ω and λ are the angular frequency and the wavelength in vacuum, respectively. $n_\parallel^*=n_\parallel+ik_\parallel$ ($n_\perp^*=n_\perp+ik_\perp$) is the complex refractive index for light polarized parallel (perpendicular) to the stress axis. n is the real refractive index and k is the attenuation index called the extinction coefficient. Equation (1) represents a wave traveling in the x direction with velocity c/n (c is the light velocity in vacuum) which is attenuated by $\exp(-2\pi kx/\lambda)$. In the piezobirefringence experiments, one measures the phase difference between the components of the light polarized parallel and perpendicular to the stress axis. The phase difference per unit path length (d) is given by[8]

$$\Delta/d=2\pi(n_\parallel-n_\perp)/\lambda\,. \tag{2}$$

In the [001] stress direction, for example, the photoelastic constant ($p_{11}-p_{12}$) or the piezo-optical constant $(\pi_{11}-\pi_{12})$ is related to $n_{\parallel}-n_{\perp}$ through

$$\Delta n \equiv n_{\parallel}-n_{\perp} = -\tfrac{1}{2}n_0^3(S_{11}-S_{12})(p_{11}-p_{12})X \tag{3}$$

or

$$\Delta n \equiv n_{\parallel}-n_{\perp} = -\tfrac{1}{2}n_0^3(\pi_{11}-\pi_{12})X\,, \tag{4}$$

where n_0 is the refractive index at zero stress, S_{11} and S_{12} are the components of the elastic compliance tensor, and X is the applied stress. The optical constants n and k are real and positive numbers and can be determined by optical measurements. They are related to the dielectric constant $(\epsilon=\epsilon_1+i\epsilon_2)$ by the following equations:

$$\epsilon_1 = n^2-k^2\,, \tag{5a}$$

$$\epsilon_2 = 2nk\,. \tag{5b}$$

The change in the refractive index Δn can now be given by (first order in stress)

$$\begin{aligned}\Delta n = \frac{\partial n}{\partial X}X &= \alpha_r(\epsilon_1,\epsilon_2)\frac{\partial\epsilon_1}{\partial X}X+\beta_i(\epsilon_1,\epsilon_2)\frac{\partial\epsilon_2}{\partial X}X\\ &= \alpha_r(\epsilon_1,\epsilon_2)\Delta\epsilon_1+\beta_i(\epsilon_1,\epsilon_2)\Delta\epsilon_2\,,\end{aligned} \tag{6}$$

with

$$\alpha_r = \frac{\partial n}{\partial \epsilon_1} = \frac{1}{4}\left(\frac{\epsilon_1+(\epsilon_1^2+\epsilon_2^2)^{1/2}}{2}\right)^{-1/2}\left[1+(\epsilon_1^2+\epsilon_2^2)^{-1/2}\epsilon_1\right], \tag{7a}$$

$$\beta_i = \frac{\partial n}{\partial \epsilon_2} = \frac{1}{4}\left(\frac{\epsilon_1+(\epsilon_1^2+\epsilon_2^2)^{1/2}}{2}\right)^{-1/2}\left[(\epsilon_1^2+\epsilon_2^2)^{-1/2}\epsilon_2\right]. \tag{7b}$$

The first and second terms of Eq. (6) are contributions from the stress-induced changes in the real and imaginary parts of the dielectric constant, respectively. The coefficients α_r and β_i are functions of photon energy, and their sign and relative magnitude determine the fractional contributions of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ to the piezobirefringence effect.

The change in the real part of the dielectric constant with applied stress can be expressed by (E_0-gap contribution)[1]

$$\Delta\epsilon_1 = \frac{\partial\epsilon_1}{\partial X}X = \sum_{i=A,B,C}\left(\frac{\partial\epsilon_1}{\partial M_i}\Delta M_i+\frac{\partial\epsilon_1}{\partial E_{gi}}\Delta E_{gi}\right), \tag{8}$$

where $M=|\langle|p|\rangle|^2$ is the squared p-matrix element, E_{gi} is the band-gap energy, and the summation indicates that contributions from the three valence bands (A, B, and C) must be included. The stress-induced change in the imaginary part of the dielectric constant, $\Delta\epsilon_2$, is also given by replacing ϵ_1 of Eq. (8) by ϵ_2.

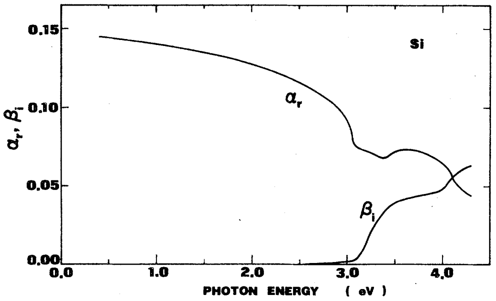

FIG. 1. The fractional coefficients α_r and β_i in Eq. (6) for Si.

III. RESULTS AND DISCUSSION

First, we consider the contribution from the imaginary part of the dielectric constant to the piezo-optical constant of Si in the region of the E_1 transition (~3.3 eV) where the sample is opaque. Figure 1 shows the spectral dependence of the fractional coefficients α_r and β_i for Si, calculated from experimental values of the optical constants reported by Philipp and Ehrenreich.[9] A low-energy region (below 3.0 eV), which contains the fundamental absorption edge, is dominated by α_r. The α_r dominance in this region makes it easy to analyze the piezobirefringence effect using a conventional technique which employs transmission of light through the sample.[8] The fractional coefficient β_i increases at photon energies above 3.0 eV, and the rising β_i and falling α_r produce a crossover at about 4.1 eV. In this region, the piezobirefringence analysis becomes very difficult because the fractions of the contributions from $\Delta\epsilon_1$ and $\Delta\epsilon_2$ should be exactly taken into account. Figure 2 shows the changes in the dielectric con-

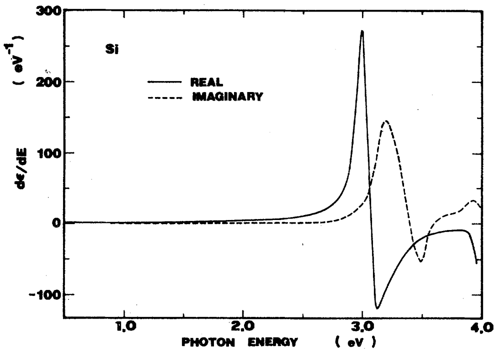

FIG. 2. The stress-induced changes in the real $\Delta\epsilon_1$ (solid line) and imaginary part $\Delta\epsilon_2$ (dashed line) of the dielectric constant for Si. The curves were obtained by numerically differentiating experimental data of the dielectric constant with respect to the photon energy.

stant $\Delta\epsilon_1$ and $\Delta\epsilon_2$ as functions of photon energy. In order to calculate $\Delta\epsilon_1$ and $\Delta\epsilon_2$ [Eq. (8)], we have used the following approximations:

$$\Delta\epsilon_1 = \frac{\partial\epsilon_1}{\partial X}X \simeq \frac{\partial\epsilon_1}{\partial E_g}\Delta E_g \simeq -\frac{\partial\epsilon_1}{\partial E}\Delta E_g \qquad (9)$$

and

$$\Delta\epsilon_2 = \frac{\partial\epsilon_2}{\partial X}X \simeq \frac{\partial\epsilon_2}{\partial E_g}\Delta E_g \simeq -\frac{\partial\epsilon_2}{\partial E}\Delta E_g\,, \qquad (10)$$

where E_g is the E_1-gap energy and E is the incident-photon energy. Such approximations have been used by Cardona and co-workers[10-13] to analyze their Raman-scattering data. We have calculated the first derivatives of ϵ_1 and ϵ_2 with respect to E according to Eqs. (9) and (10) by numerically differentiating experimental data of Philipp and Ehrenreich.[9] The obtained results are shown in Fig. 2 by solid ($\Delta\epsilon_1$) and dashed lines ($\Delta\epsilon_2$). The changes in the dielectric properties under strain have also been calculated by Tsay, Bendow, and Mitra[14] in terms of a full band-structure approach to estimate the Brillouin-scattering efficiencies of Ge and Si. Piezobirefringence measurements in the opaque region have been recently reported in Si,[5] and GaP,[6] along with the new method employing the Raman-scattering technique as a probe. Now we compare our theoretical model with the data on Si.[5] Figure 3 shows the theoretical curve calculated from Eq. (6) along with the experimental data (Fig. 7 of Ref. 5). The data below 1.5 eV are obtained from Ref. 8 (conventional method) and those above 1.5 eV from Ref. 5 (new method). The filled circles are plotted on the scale indicated in the figure, while the open circles are replots of the same values on an expanded (4×) scale. The theoretical curve was calculated using the fractional coefficients and the changes in the dielectric constant given in Figs. 1 and 2, respectively. It is evident from Fig. 3 that the experiment and calculation are in quite good agreement.

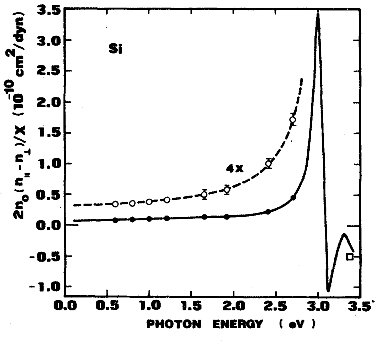

FIG. 3. The theoretical dispersion of the piezo-optical constant for Si calculated from Eq. (6). The experimental data are obtained form Ref. 5. The filled circles are plotted on the scale indicated in the figure, while the open circles are replots of the same values on an expanded (4×) scale and the square is plot of the data at $E = 3.38$ eV on a reduced ($\frac{1}{10}$×) scale.

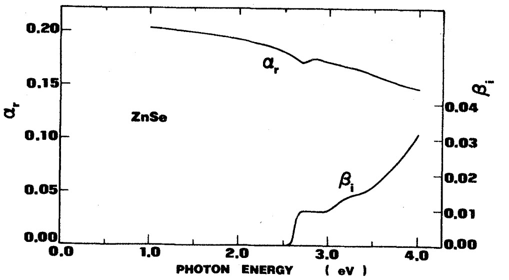

FIG. 4. The fractional coefficients α_r and β_i in Eq. (6) for ZnSe.

Chandrasekhar, Grimsditch, and Cardona[5] also reported the experimental value at $E = 3.38$ eV in Si measured by their new method. The square in Fig. 3 is the data point at $E = 3.38$ eV, where it is plotted on a reduced ($\frac{1}{10}$×) scale. Due to the lack of points at intermediate photon energies they have not been able to infer the sign of the effect at $E = 3.38$ eV. They have suggested that in the photon-energy region close to the critical point real transitions occur and the exact linewidth, including the imaginary part of the stress-induced dielectric constant, is required for an accurate description of the piezobirefringence phenomena. By virtue of the present model, we can estimate the sign of the data at $E = 3.38$ eV to be negative because the signs of α_r and β_i are positive in this photon-energy region but those of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ are negative.

Next we consider the contribution from the imaginary part of the dielectric constant to the piezobirefringence coefficient in ZnSe determined from the resonant-Brillouin-scattering experiments.[1] Figure 4 shows the spectral dependence of the fractional coefficients α_r and β_i for ZnSe, calculated from experimental values of the optical constants reported by Aven, Marple, and Segall.[15] The stress-induced changes in the dielectric constant, $\Delta\epsilon_1$ and $\Delta\epsilon_2$, as functions of wavelength for the case of [001] stress direction are shown in Fig. 5 by the solid ($\Delta\epsilon_1$) and dashed line ($\Delta\epsilon_2$). In the calculations, we have replaced ω by $\omega + i(\Gamma/\hbar)$ in Eq. (13) [or Eq. (14)] of Ref. 1 and calculated the real and imaginary parts of this equation using the numerical parameters given in Ref. 1, where ω is the angular frequency of the light and Γ is the lifetime-broadening energy. It is important to point out that the change in the imaginary part ($\Delta\epsilon_2$) has considerably large value

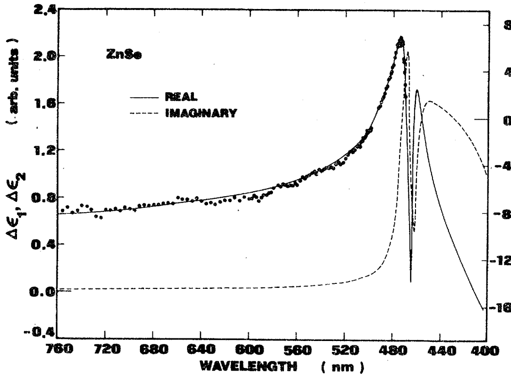

FIG. 5. The stress-induced changes in the real $\Delta\epsilon_1$ (solid line) and imaginary part $\Delta\epsilon_2$ (dashed line) of the dielectric constant for ZnSe along with the experimental data on the photoelastic constant $p_{11}-p_{12}$.

in the region very close to the fundamental absorption edge (2.68 eV$\simeq$463 nm). However, the fractional coefficient β_i is very small in the photon-energy region compared with α_r. It is clear from this fact that the contribution from the imaginary part of the dielectric constant to the piezobirefringence effect is much smaller than that from the real part of the dielectric constant, and therefore we can disregard the imaginary-part contribution for ZnSe in the region below and near the lowest direct gap in a good approximation. From these considerations, the photoelastic constant $p_{11}-p_{12}$ becomes proportional to $\alpha_r\Delta\epsilon_1$ [see Eqs. (3) and (6)]. We have, therefore, fitted our experimental data[1] on the photoelastic constant $p_{11}-p_{12}$ to the calculated curve (solid line) by taking into account the constant term which arises from the higher-gap contributions (i.e., E_1, $E_1+\Delta_1$, and E_2 transitions), where we have assumed that α_r is nondispersive in the measured wavelength region (see Fig. 4). The plotted data points are obtained from the Brillouin-scattering measurement. (Note that the Brillouin-scattering intensity is proportional to the square of the photoelastic constant.)[16,17] It is clear from the figure that the calculated curve shows an excellent agreement with the experimental data. From these discussions, we conclude that the photoelastic constants in the region of transparency can be safely determined only by the stress-induced change in the real part of the dielectric constant, and therefore our previous results[1-3] can be used without any modification.

It can be found from Figs. 1 and 4 that the spectral dependence of the fractional coefficients α_r and β_i is very similar to that for the modulated reflectance spectroscopy proposed by Seraphin and Bottka[18]:

$$\Delta R/R=\alpha(\epsilon_1,\epsilon_2)\Delta\epsilon_1+\beta(\epsilon_1,\epsilon_2)\Delta\epsilon_2\,, \tag{11}$$

where the coefficients α and β, which are obtained by differentiating Fresnel's formula, are functions of photon energy, and their sign and relative magnitude determine the fractional contributions of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ to the modulation spectroscopy. From the spectral dependence of ϵ_1 and ϵ_2 for semiconductors such as II-VI compounds, we have $\alpha\gg\beta$ in the region of the fundamental absorption edge, i.e., only the first term of Eq. (11) is important, and therefore $\Delta R/R$ is determined predominantly by the nature of the fraction $\Delta\epsilon_1$. In the case of piezoreflectance spectroscopy,[19] the change in the dielectric constant $\Delta\epsilon_1$ in Eq. (11) is given by the same expression as Eq. (8). We recently studied a relation between resonant Brillouin scattering (piezobirefringence) and first-derivative modulation spectroscopy such as piezoreflectance, thermoreflectance, and wavelength-derivative spectroscopy, and compared the experimental Brillouin spectra with the first-derivative modulation spectra obtained in some semiconductors.[7] The obtained results have clearly suggested a close relationship between them. This is of course to be expected from the expressions of Eqs. (6) and (11).

In conclusion, we have obtained a generalized expression of the piezobirefringence effect by taking into account both the stress-induced changes in the real ($\Delta\epsilon_1$) and imaginary part ($\Delta\epsilon_2$) of the dielectric constant. The coefficients α_r and β_i, which are functions of photon energy, have been calculated from an analytical point of view. Such coefficients determine the fractional contributions of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ to the piezobirefringence response. The present model has been demonstrated for Si and ZnSe in the opaque photon-energy region (E_1 edge) and in the transparency region (E_0 edge), respectively. Good agreement between the experiment and calculation has been found. When the present method is adopted, it is possible to extend the piezobirefringence analysis to a large number of opaque materials.

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