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Frequency dependence of the photoelastic coefficients of silicon

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We have measured the frequency dependence of the individual photoelastic coefficients of silicon using a simplified acousto-optic technique. We define the photoelastic tensor q by $\Delta\epsilon_{ij} = q_{ijkl}e_{kl}$, where e is the particle displacement gradient tensor and $\Delta\epsilon$ is the strain-induced change in the optical dielectric tensor. We find $q_{1111}(3.39\ \mu\text{m}) = +13.0 \pm 0.6$, $q_{1111}(1.15\ \mu\text{m}) = +15.7 \pm 1.1$, $q_{1122}(3.39\ \mu\text{m}) = -2.41 \pm 0.1$, and $q_{1122}(1.15\ \mu\text{m}) = -1.45 \pm 0.1$. The extrapolated long-wavelength limit of the average photoelastic coefficient agrees well with our previous estimate for the frequency-independent Phillips–Van Vechten model. However, within a proper frequency-dependent Penn model we have shown that the "oscillator strength" does not vary as r^{-3}. Further we have demonstrated that the dispersion energy E_d in the Wemple-DiDomenico model is proportional to $r^{-1.9 \pm 0.8}$ and not volume independent as would be expected from the model. It is concluded that whereas a simple single-gap model works well to describe the low-frequency dispersion in the dielectric constant of silicon, it is incapable of describing the dispersion in the photoelastic tensor.

I. INTRODUCTION

There are numerous microsopic or phenomenological models describing physical many-body systems. The interaction of electromagnetic radiation with the system is described by the model dielectric response function $\epsilon(\vec{r}, t)$ or its Fourier transform $\epsilon(\vec{q}, \omega)$. The average $(q \to 0)$ dielectric tensor $\epsilon(\omega)$ is frequently compared with experiments to check or determine model parameters. When a long-wavelength particle displacement gradient exists in the system, ϵ changes to $\epsilon(0, \omega) + \Delta\epsilon(0, \omega)$. The photoelastic tensor $q(\omega)$ relates strain-induced changes in ϵ to the nonsymmetrized particle displacement gradients, e;

$$\Delta\epsilon_{ij}(\omega) = q_{ijkl}(\omega)\, e_{kl}\,. \tag{1}$$

In many recent dielectric models, semiempirical assumptions are made relating various model parameters to interatomic spacing. Measurements of the strain-induced changes in ϵ can be used to test the validity of these assumptions. Measurements of the frequency dependence of q allows one to separate out the strain dependence of individual model parameters, as well as to check the dispersion predicted by explicitly frequency-dependent models. We will restrict ourselves here to only a few of the models for fourfold coordinated covalent semiconductors. In the Phillips–Van Vechten (PVV) model,[1] a universal power-law relation is assumed between the average homopolar gap and the interatomic spacing (or volume). It is implicit that the relation is valid both for intra- as well as intermaterial variation. Similarly, in the Harrison bond-orbital model,[2] a simple power law is assumed for the r dependence of V_2. In the Wemple-DiDomenico (WD) model,[3] a two-parameter one-oscillator model is used. It is found empirically that the ratio of the oscillator strength to oscillator resonant frequency is very nearly constant for covalent systems when scaled for such configurational characteristics as formal ionic valency and nearest-neighbor coordination. In particular, this "dispersion energy" should be independent of volume. Finally, we present our determination of the volume dependence of the parameters in the Chadi-White (CW) model,[4] a two-parameter tight-binding model.

We have measured the photoelastic coefficients of silicon, a prototype covalent solid, at 3.391 and 1.152 μm (just below the indirect edge). In Sec. II, we will discuss the experimental methods; in Sec. III, we will discuss the results and compare our results with other related measurements; in Sec. IV, we will discuss various models and consider the implications.

II. EXPERIMENTAL METHOD

Figure 1 shows a simplified block diagram of the apparatus used to measure the room-temperature frequency dependence of the individual photoelastic coefficients relative to the known values[5] at 3.391 μm. The apparatus is similar to that described previously in Ref. 5; only the important changes will be presented here.

Because dispersion measurements need not be absolute, no reference material was used. An ultrasonic pulse (280 MHz) launched by a 25-MHz fundamental, 3-mm square, 35° rotated Y-cut LiNbO_3 transducer propagates forth and back in the sample and diffracts the light beam during each pass. The diffracted intensity is a function of $\phi \equiv \alpha q^2 P/\lambda_0^2 \epsilon \rho v^3$, where α depends on the aspect ratio of the transducer, λ_0 is the vacuum wavelength, P is the acoustic power density, ρ and v are, respectively, the density and acoustic mode velocity of

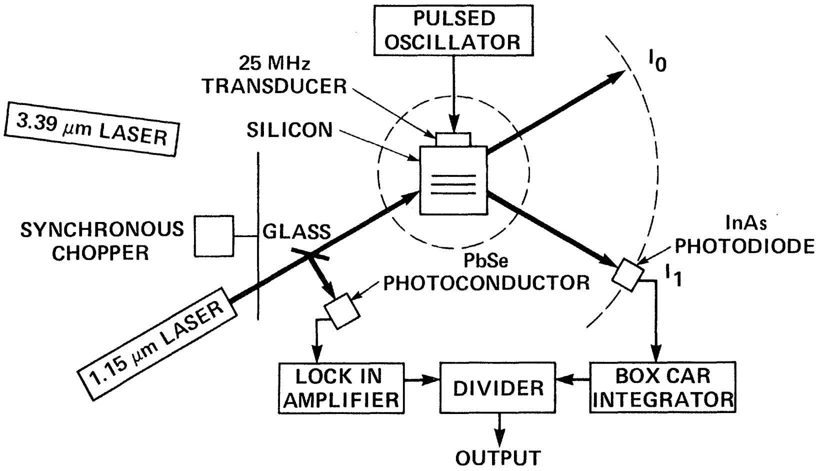

FIG. 1. Block diagram of the apparatus.

the sample. The two laser beams are transmitted through approximately the same region of the sample. However, to eliminate any effects of acoustic attenuation arising from slight misalignment, we measure the signal from the transmitted and reflected pulses to obtain an expression which is independent of the position of the light in the sample.[6] The first-order signals are normalized by dividing the intensity of the light transmitted when the sound pulse is absent. A part of the chopped incident beam is sampled by a PbSe photoconductor and lock-in amplified. The low-frequency output is sent to the B channel of a divider and normalizes the output of the boxcar integrator to account for source fluctuations. Using this technique, the worst case signals having a signal-to-noise ratio of approximately two and peak-to-peak source fluctuations of 10% were improved to better than 50 and 1%, respectively. The InAs photodiode was calibrated at the two laser wavelengths by comparison with a thermopile (in the linear response region of the thermopile). The calibration was reproducible to only ~ 3% (rms) and represents the largest source of error in the experiment.

In the Bragg diffraction regime,[7] ϕ is simply related to the normalized intensity $\hat{I}$ by

$$\phi = \sin^{-2} \hat{I} . \qquad (2)$$

As the light wavelength decreases relative to the acoustic wavelength, one can no longer use Eq. (2) to determine ϕ. We have solved the difference-differential equations for the acousto-optic interaction using a Hamming integration routine and found that the largest correction occurs for the transmitted 280-MHz pulse with the 1.15-μm laser polarized to measure q_{1111}. We find a worst-case correction to q less than 2%. The exact integration values have been used in all cases instead of (2). The geometric mean $\hat{\phi}$ of the values of ϕ obtained from the transmitted and reflected pulses is independent of position along the length of the sample. (We have used a transducer having square cross section so that the aspect ratio of the acoustic beam does not change as the pulse propagates.) Finally, then, from the ratio $\hat{\phi}(1.152)/\hat{\phi}(3.391)$, we ascertain the relative dispersion $\xi \equiv q(1.152)/q(3.391)$.

III. EXPERIMENTAL RESULTS

In Table I, we have listed the measured values of ξ_{1111} and ξ_{1122}. The error indicated is derived from the approximate rms deviation in the distribution of experimental values and the distribution in diode calibration measurements. We have also given the measured values[5] of q_{1111} and q_{1122} at 3.391 μm and the derived values at 1.152 μm [i.e., $q(1.15) = q(3.39) \times \xi$]. We have used the extrapolated-refractive-index data of Salzberg and Villa[8] throughout.

To our knowledge, these are the first measurements of the frequency dependence of the individual photoelastic tensor components of silicon. By taking linear combinations of the individual coefficients, comparisons can be made to reported values of hydrostatically induced changes in the refractive index, as well as uniaxial piezobirefringence; $\bar{q} \equiv (q_{1111} + 2q_{1122})$ and $\Delta q \equiv (q_{1111} - q_{1122})$, respectively.

To interpolate between our measurements, we utilize the fact that due to time-reversal invariance of the Hamiltonian, the photoelastic dispersion can be expressed as an expansion in $(E/E_0)^2$, where E is $\hbar\omega$ and E_0 is an average gap slightly greater than

TABLE I. Experimental results.

ξ_{1111}	ξ_{1122}	$q_{1111}(3.391)$	$q_{1111}(1.152)$
$+1.21 \pm 0.06$	$+0.60 \pm 0.03$	$+13.0 \pm 0.6$	$+15.7 \pm 1.1$
$q_{1122}(3.391)$	$q_{1122}(1.152)$	$\epsilon(3.391)$	$\epsilon(1.152)$
-2.41 ± 0.1	-1.45 ± 0.1	11.758	12.468

the lowest direct gap. In transmission experiments in silicon, the largest E is limited by the indirect absorption edge at 1.1 eV; therefore, $(E_{\max}/E_0)^2 \lesssim 0.08$. The photoelastic dispersion thus reduces to

$$q(E) = \alpha_0 + \alpha_1 (E/E_0)^2 . \tag{3}$$

From our measurements, we find $\alpha_{0,1111} = 12.6 \pm 0.6$, $\alpha_{1,1111}/E_0^2 = 2.63 \pm 0.3$, $\alpha_{0,1122} = -2.54 \pm 0.1$, and $\alpha_{1,1122}/E_0^2 = 0.94 \pm 0.09$.

From the most recent and accurate hydrostatic measurements of $d\ln n/dV$, Vetter[9] estimates that $\bar{q}$ increases by 32% from 2.5 to 1.1 μm. From our results, the interpolation indicates that $\bar{q}$ increases by 55% in the same range.

Using the elastic constant measurements of McSkimin and Andreatch,[10] we have calculated Δq from the measurements of Nikitenko and Martynenko[11] and of Higginbotham, Cardona, and Pollak.[12] Nikitenko *et al.* found a *decrease* of 10% from 1.4 to 1.1 μm. In the more recent measurements of Higginbotham *et al.*, as well as the present measurements, there is an *increase* of 6% in the same range and a 10% increase from 2.02 to 1.1 μm. We therefore believe that our data for the frequency dependence of the photoelastic coefficients agree well with the most accurate related data.

IV. THEORETICAL IMPLICATIONS

The photoelastic coefficients are a measure of the strain dependence of ϵ. In principle they can therefore be used to check or determine the strain dependence of parameters in models for which ϵ can be calculated. We will relate our measurements to several currently popular simple models of covalent systems. In particular, we will consider here the PVV model,[1] the Harrison bond-orbital model,[2] a frequency-dependent extension of the Penn model,[13–15] the WD model,[3] and the CW model.[4] Only the latter three models listed have an explicit frequency dependence.

To treat the former theories, we must use the value of q extrapolated to $E = 0$; i.e., $q(0) = \alpha_0$. In a previous paper,[5] we made the assumption that the zero-frequency limit of the average photoelastic coefficient $\bar{p} = -\bar{q}/\epsilon^2$ of silicon can be well approximated by the value measured at 3.39 μm (0.366 eV). That the residual dispersion is small is reasonable because 0.366 eV is an order of magnitude smaller than the lowest direct gap. One purpose of the work reported here was to determine $\bar{q}(0)$ to validate our assumption.

In the Penn-PVV model,

$$\epsilon(0) - 1 = (E_P^2/E_0^2)(1 - \Delta + \tfrac{1}{3}\Delta^2) , \tag{4}$$

where E_P is the plasma energy of the valence electrons, $\Delta = E_0/4E_F$ and E_F are the calculated Fermi energies of the free-electron gas, and

$$\bar{q}(0) = [\epsilon(0) - 1](2s - 3) , \tag{5}$$

where $s \equiv -d\ln E_0/d\ln r$. Our experimental value of $\bar{q}(0) = 7.6 \pm 0.4$ implies that $s = 1.86 \pm 0.02$, which is, in fact, in close agreement with our earlier estimate.

In a similar manner, within the Harrison bond-orbital model,

$$\bar{q}(0) = [\epsilon(0) - 1]\{[d\ln(\gamma^2/V_2)/d\ln r] - 1\} , \tag{6}$$

where V_2 is the "covalent energy," and γ is a bond-dipolar scaling parameter. The measured value of $\bar{q}(0)$ implies that $d\ln(\gamma^2/V_2)/d\ln r = 1.71 \pm 0.03$. If Harrison's assumption that $d\ln V_2/d\ln r = -2$ is correct, then the measurements imply a very small logarithmic derivative of γ (i.e., γ is nearly independent of nearest-neighbor separation).

In fact, we can be more explicit in treating the Penn-PVV model. Martin[14] and Breckenridge, Shaw, and Sher,[15] using the proper momentum-dependent matrix elements, have derived an expression within the Penn model for the frequency dependence of ϵ in the limit $q \to 0$. For $E/E_0 \ll 1$, we find (to order Δ^2)

$$\epsilon(E) - 1 = \frac{2}{3}\frac{E_P^2}{E_0^2}\Big[(1 - \Delta + \tfrac{1}{2}\Delta^2) + \tfrac{4}{5}(1 - \tfrac{3}{4}\Delta + \tfrac{1}{4}\Delta^2) \times \left(\frac{E}{E_0}\right)^2 + \tfrac{24}{35}(1 - \tfrac{5}{8}\Delta + \tfrac{1}{6}\Delta^2)\left(\frac{E}{E_0}\right)^4 + \cdots\Big] . \tag{7}$$

[The factor of $\frac{2}{3}$, which is not present in the Penn-PVV dielectric function, does not change the logarithmic derivative of $(\epsilon - 1)$ and therefore does not affect the derivation of $d\ln E_0/d\ln r$ from photoelastic measurements; nor does it significantly alter the PVV result for the *inter*-material variation of E_0 with r.] Equation (7), a one-parameter model, can be compared with a polynomial fit to the refractive-index data of Ref. 8. A fit to the experimental data gives $\epsilon(E) - 1 = \beta_0 + \beta_1(E/E_0)^2 + \beta_2(E/E_0)^4$, with $\beta_0 = 10.67$, $\beta_1/E_0^2 = 0.628$, and $\beta_2/E_0^4 = 0.051$. The extended Penn model gives $E_0 = 3.99$ eV and $\beta_0 = 10.67$ (by fit), $\beta_1/E_0^2 = 0.545$, and $\beta_2/E_0^4 = 0.034$. The low-frequency dispersion predicted by the model is thus substantially smaller than the data.

The hydrostatic photoelastic coefficient $\bar{q}$ in the extended Penn model is given by

$$\bar{q}(E) = \frac{2}{3}\frac{E_P^2}{E_0^2}\{[(1 - \Delta) + (1 - \tfrac{3}{4}\Delta)\Omega^2]F' + [(2 - \Delta) + (4 - \tfrac{9}{4}\Delta)\Omega^2]s - (2\Delta + \tfrac{3}{2}\Delta\Omega^2)\} , \tag{8}$$

where $F' \equiv d\ln E_P^2/d\ln r$, and $\Omega^2 \equiv \frac{4}{5}E^2/E_0^2$. If we let E_P^2 be an empirical parameter in the model and determine the r dependence of E_P^2 from fitting the frequency dependence of $\bar{q}$, we find $F' = -6.9 \pm 0.9$

instead of -3, and $s = 3.7 \pm 0.7$ instead of 1.86.

The second explicit frequency-dependent model considered here is the empirical dielectric function of WD. They find that a single two-parameter Sellmeier oscillator

$$\epsilon(E) - 1 = F/E_0^2 - E^2 \tag{9}$$

fits the dielectric dispersion of most materials for which the dispersion is known. For silicon, we find $F = 178.8$ eV^2 and $E_0 = 4.09$ eV. Fitting with a polynomial fourth order in (E/E_0), we obtain $\beta_0 = 10.67$, $\beta_1/E_0^2 = 0.634$, and $\beta_2/E_0^4 = 0.043$ in much better agreement with the data than is the Penn model. The photoelastic coefficients derived from the WD model are

$$q(E) = \left(\frac{F}{E_0^2 - E^2}\right)\left[\frac{d\ln F}{d\ln x} - 2\left(\frac{E_0^2}{E_0^2 - E^2}\right)\frac{d\ln E_0}{d\ln x}\right], \tag{10}$$

where $d\ln x$ is either $d\ln r$ in the hydrostatic case or e_{11} in the uniaxial case. For this model, we find again that if the oscillator strength is proportional to r^{-3}, then $s = 1.86$. If, however, we let the variation of the oscillator strength be a free parameter (in keeping with the model), we find $s = 2.6 \pm 0.5$ and $F' = -4.5 \pm 0.6$. Moreover, WD have demonstrated that $E_d = F/E_0$ is constant within classes of materials having the same nearest-neighbor cation coordination number, anion valency, and effective number of valence electrons per anion. We would therefore expect E_d to be independent of volume because scaling does not change the configurational class of the material. However, we find here that E_d is not independent of r. In fact, we find $E_d \alpha r^{-1.9 \pm 0.8}$.

The CW model[4] is a tight-binding two-band model for covalent semiconductors, which is solved in terms of a Bethe lattice. For $E \ll E_0$, the Bethe lattice sums include only nearest contributions and should approximate well the real lattice sums. The CW model gives for $E < E_0$

$$\epsilon(E) - 1 = \tfrac{3}{2}(E_P^2/E_0)\{[\Delta_- + 2(\Delta_-^2 - 12V_1^2)^{1/2}]^{-1} + [\Delta_+ + 2(\Delta_+^2 - 12V_1^2)^{1/2}]^{-1}\}, \tag{11}$$

where E_0 is the energy gap, $\Delta_\pm = E_0 \pm E$ and V_1 is equal to the difference in the valence and conduction band widths. The model is a two-parameter model which satisfies the oscillator sum rule. For this model, the low-frequency dielectric dispersion $\beta_0 = 10.67$, $\beta_1/E_0^2 = 0.615$, and $\beta_2/E_0^4 = 0.063$, is in quite good agreement with the experimental data. From a comparison of $d\ln[\epsilon(E) - 1]/d\ln r$ in the model with the measured values of $\bar{q}$, we find $s = 1.70 \pm 0.13$ and $d\ln V_1/d\ln r = 0.14 \pm 0.6$. These results imply that within the model, the volume dependence of the direct gap is similar to that of the average gap in the PVV model and that V_1 is almost independent of volume.

In the case of silicon, the photoelastic dispersion in the region accessible using the present technique is not very large compared with the experimental uncertainty. This arises from the fact that in silicon, the lowest-absorption edge lies far below the average gap E_0. Using either frequency-dependent model discussed above, we can determine the percentage variability in the derived quantities $d\ln s$ and $d\ln F'$ or $d\ln V_1$ arising from possible errors in the absolute measurements of q at 3.391 μm and the relative dispersion measurements at the two optical wavelengths. Taking the variability in parameter X as

$$|d\ln X| = \left[\left(\frac{d\ln X}{d\ln q(3.39)}\right)^2 [d\ln q(3.39)]^2 + \left(\frac{d\ln X}{d\ln \xi}\right)^2 (d\ln \xi)^2\right]^{1/2}, \tag{12}$$

we find with $|d\ln q|$ and $|d\ln \xi| \simeq 0.05$ at both wavelengths, that for the extended Penn model and the WD model $|d\ln s| \sim 0.18$, and $|d\ln F'| \sim 0.13$. For the CW model, $|d\ln s| \sim 0.08$, and $|d\ln V_1'| \sim 4.0$. Unfortunately, the uncertainty in the determination of the parameters is large. Nevertheless, in neither the extended Penn model nor the WD model is an inverse volume dependence found for the oscillator strength within the experimental error.

V. CONCLUSION

We have presented here the results of our measurements of the dispersion in the photoelastic tensor of silicon. We have determined the valence dependence of model parameters in the Harrison bond-orbital model and the CW model. We find that the extrapolated low-frequency limit of the average photoelastic coefficient agrees well with our previous estimate for the frequency-independent PVV model. On the other hand, we have determined an expression for the average photoelastic coefficient in a frequency-dependent Penn model and shown that the "oscillator strength" does not vary as r^{-3}. Finally, we have shown that, at least in silicon, the WD dispersion energy is not independent of interatomic spacing, as would be expected from the model. It is concluded that for the single-gap models discussed, the intramaterial volume dependence differs from the explicit intermaterial scaling.

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[1] J. A. Van Vechten, Phys. Rev. 182, 891 (1969).

[2] W. A. Harrison and S. Ciraci, Phys. Rev. B 10, 1516 (1974).

[3] S. H. Wemple and M. DiDomenico, Jr., Phys. Rev. B 1, 193 (1970).

[4] D. J. Chadi and Robert M. White, Phys. Rev. B (to be published).

[5] David K. Biegelsen, Phys. Rev. Lett. 32, 1196 (1974).

[6] R. W. Dixon and M. G. Cohen, Appl. Phys. Lett. 8, 205 (1966).

[7] W. R. Klein and Bill D. Cook, IEEE Trans. Sonics Ultrason. SU-14, 123 (1967).

[8] C. D. Salzberg and J. J. Villa, J. Opt. Soc. Am. 47, 244 (1957).

[9] R. Vetter, Phys. Status Solidi A 8, 443 (1971).

[10] H. J. McSkimin and P. Andreatch, Jr., J. Appl. Phys. 35, 2161 (1964).

[11] V. I. Nikitenko and G. P. Martynenko, Fiz. Tverd. Tela 7, 622 (1965) [Sov. Phys.-Solid State 7, 494 (1965)].

[12] C. W. Higginbotham, M. Cardona, and F. H. Pollak, Phys. Rev. 184, 821 (1969).

[13] D. Penn, Phys. Rev. 128, 2093 (1962).

[14] R. M. Martin (private communication).

[15] Roger A. Breckenridge, Robert W. Shaw, Jr., and Arden Sher, Phys. Rev. B 10, 2483 (1974).