

Table II. Properties of He³ at the critical point.

$T_c = 3.324 \pm 0.0018^\circ\text{K}^a$
$p_c = 873.0 \pm 1.5 \text{ mm}^a$
$\rho_c = 0.0418 \pm 0.001 \text{ g cm}^{-3}$

^aReference 2.

at the critical point are summarized.

From the present isochore data the critical-point exponents for compressibility, γ_g' , γ , and γ_l' have also been derived. The values found are

$$\gamma_g' = 1.00 \pm 0.05,$$

$$\gamma = 1.09 \pm 0.05,$$

$$\gamma_l' = 1.18 \pm 0.10,$$

in the relationship

$$\kappa_T \propto |T_c - T|^{-\gamma}.$$

γ_g' and γ_l' refer to the vapor and liquid branches of the coexistence curve, and γ refers to the critical isochore for $T > T_c$. While β is derived directly from density observations, exponents such as γ are derived from derivatives of PVT data and hence are subject to more uncertainty.

An interpolation of the isochore data has been carried out at the critical temperature, 3.324°K , and the degree of the critical isotherm, δ , was evaluated in the relationship

$$|P - P_c| \propto |\rho - \rho_c|^\delta.$$

The value of δ found was 3.4 ± 0.2 , which is

quite different from the value 4.2 obtained by Widom and Rice⁹⁻¹¹ for a variety of other substances. A discussion of the reasons for this deviation in δ as well as that found for β , in terms of quantum mechanical corrections to the theory of corresponding states, has been given by Sherman and Hammel.¹²

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⁵For the critical-point exponents the notation of M. E. Fisher, *J. Math. Phys.* **5**, 944 (1964), is used.

⁶E. C. Kerr, *Phys. Rev.* **96**, 551 (1954).

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⁸B. Widom and O. K. Rice, *J. Chem. Phys.* **23**, 1250 (1955).

⁹S. Y. Larsen and J. M. H. Levelt Sengers, in *Advances in Thermophysical Properties at Extreme Temperatures and Pressures*, edited by S. Gratch (American Society of Mechanical Engineers, New York, 1965), p. 74. Larsen and Sengers point out that the value of δ determined by Widom and Rice for classical systems is likely to be too small.

¹⁰R. H. Sherman and E. F. Hammel, *Phys. Rev. Letters* **15**, 9 (1965).

DEPENDENCE OF THE OPTICAL CONSTANTS OF SILICON ON UNIAXIAL STRESS

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Structure in the real and imaginary parts of the dielectric constant, ϵ_1 and ϵ_2 , is well known to be related to the presence of critical points in the (direct) optical energy versus k -vector relation.¹ These critical points have played a major role in studies of band structure.² Recently Seraphin³ has been able to modulate the reflectivity by an ac variation of the electric field at the surface. This modulation

greatly enhances the structure and hence makes it possible to resolve critical points much more clearly than in earlier methods.

We wish to report here an alternative method for enhancing critical-point structure, namely by an ac strain modulation of the reflectivity, and to present a simple theory of the effect and the symmetry relations between the strain components and the polarization direction of

the reflected light. We find that the strain modulation can be represented by three fundamental constants which may be used to characterize and, in special cases, to infer the symmetry properties of the critical points. This three-parameter characterization of critical points should be extremely useful in comparing critical points in different materials and identifying corresponding points.

The theory of the strain effect is very simply related to the deformation-potential constants of the critical-point energies.⁴ The strain variation takes a derivative of the square-root singularities in ϵ_1 and ϵ_2 leading to 1/square-root singularities which are infinite (in the absence of lifetime broadening) at the critical points, thus greatly enhancing the structure.

Dc measurements of piezoreflectance have been made by Philipp, Dash, and Ehrenreich⁵ and by Gerhardt.⁶ Because the reflectivity changes are very small, even for strains of the order of the elastic limit of silicon ($\Delta L/L \sim 10^{-2}$),⁷ the dc strain method is rather insensitive. The sensitivity can be considerably improved by use of phase- and frequency-sensitive detection of ΔR .

In this work, the stress is applied to the crystal via the method of free-free longitudinal acoustic resonance, and the experimental apparatus is diagrammed in Fig. 1. A $+5^\circ X$ -cut extensional quartz-crystal oscillator trans-

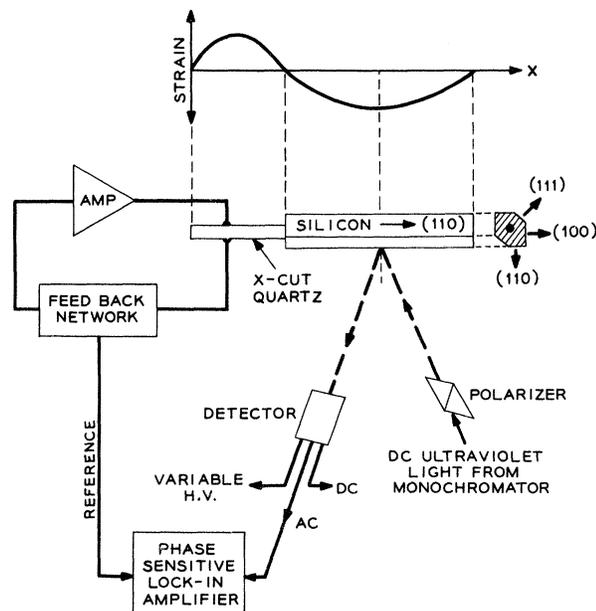


FIG. 1. Diagram of the ac piezoreflectance experiment.

ducer is cemented to a thin Si bar. The free longitudinal resonance of the quartz driver is $f_0 \sim 130$ Kc/sec, and the silicon-crystal length is carefully ground and (finally) etched so that the composite oscillator system has the resonance frequency $f_0 \pm 0.5$ cps. Under the assumption that the quartz and silicon crystals are thus tuned to the same fundamental longitudinal resonance, the bond interface between the two crystals is a position of maximum vibrational amplitude and of minimum (or zero) stress. Confirmation of this assumption comes from the fact that the computed resonance length agrees with the measured length to better than 1%. The amplitude limit of vibration is determined only by the elastic limit of the "weaker" of the two components, in this case the quartz, which has the limit $\Delta L/L \sim 5 \times 10^{-4}$. Monochromatic dc light is reflected from the midpoint of the Si-crystal length (point of maximum stress), and this light is detected by a uv-sensitive photomultiplier tube (response time $\leq 10^{-8}$ sec).

The output of the photomultiplier tube is monitored by a dc current meter. This current is proportional to the total reflectivity R . The ac (130-Kc/sec) photomultiplier output is fed into a phase-sensitive lock-in detector which uses a small fraction of the quartz-crystal driver circuit as a reference signal. This demodulated signal is proportional to ΔR , and thus $I(\omega)/I_{dc} = \Delta R/R$, which is the quantity which is plotted. Results of measurements of $\Delta R(\omega)/R(\omega)$ for polarizations parallel and perpendicular to the stress direction for the 001, $1\bar{1}0$, and $1\bar{1}\bar{1}$ faces with 110 stress and for an 010 face with 100 stress are shown in Fig. 2 and are labeled according to the polarization direction of the E vector. In the case of 110 stress, 110 polarization measurements were made on all three principal faces. Since the stress is longitudinal for a long thin bar, the reflectance should be the same for all faces for 110 polarization. The data shown in Fig. 2 demonstrate clearly that such is the case, save for a modest wavelength-insensitive constant difference between curves for the various faces. On the other hand, the dependences of $\Delta R/R$ on $h\nu$ for polarizations with \vec{E} vector perpendicular to the stress show major variations from one another and from the parallel polarization curves.

For small strains the change in reflectivity can be related directly to a variation in the dielectric-constant tensor, ϵ_{ij} , proportional

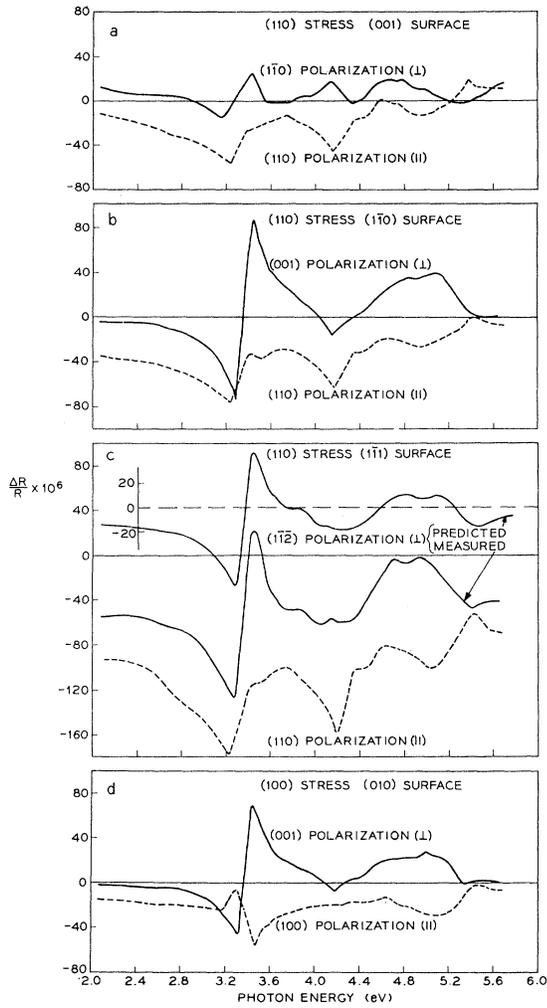


FIG. 2. $\Delta R/R$ vs $h\nu$ for polarization modes parallel and perpendicular to stress field.

to the strain u_{kl} . The constant of proportionality is a fourth-order tensor, with W defined, for the case of cubic symmetry with cubic axes, by

$$\begin{aligned}\Delta\epsilon_{xx} &= W_{11}u_{xx} + W_{12}u_{yy}, \\ \Delta\epsilon_{xy} &= W_{44}u_{xy}.\end{aligned}\quad (1)$$

We adopt the abbreviations which are used for the stress-strain tensor, c .

The change in reflectance, ΔR , for a cubic material can be shown to be given by

$$\begin{aligned}\Delta R &= \frac{2}{[(n+1)^2 + k^2]^2(n^2 + k^2)} \\ &\times \text{Re}\{[(n-ik)^3 - (n-ik)]\Delta\epsilon_{ii}\}.\end{aligned}\quad (2)$$

Re means "real part of." $\Delta\epsilon_{ii}$ is the diagonal

component of the dielectric-constant tensor in the direction of polarization of the E vector of the light. n and k are the optical constants of the unstrained (cubic) material which are therefore scalars.

In a long thin bar the stress tensor, T_{ij} , will be entirely diagonal (longitudinal), T_{jj} , where j is parallel to the length of the bar. For a bar of finite thickness the corrections to this result go as $(w/l)^2$, where w/l is the width to length ratio. This correction is of the order of 1% in this experiment and will henceforth be neglected. For a (110) bar ($T_{jj} \equiv T_{110}$), we have

$$T_{xx} = T_{yy} = T_{xy} = \frac{1}{2}T_{110}, \quad (3)$$

where T_{xx} , etc., are the stresses resolved along cubic directions. The elastic-compliance constants, s_{ij} , may then be used with (1) and (3) to give the relations

$$\begin{aligned}\epsilon_{110}^{110} &= [W_{11}(s_{11} + s_{12}) + W_{12}(s_{11} + 3s_{12}) \\ &\quad + W_{44}s_{44}] \frac{1}{2}T_{110},\end{aligned}\quad (4)$$

$$\begin{aligned}\epsilon_{110}^{110} &= [W_{11}(s_{11} + s_{12}) + W_{12}(s_{11} + 3s_{12}) \\ &\quad - W_{44}s_{44}] \frac{1}{2}T_{110},\end{aligned}\quad (5)$$

$$\epsilon_{001}^{110} = [2W_{11}s_{12} + 2W_{12}(s_{11} + s_{12})] \frac{1}{2}T_{110}. \quad (6)$$

We can also derive the relation

$$\epsilon_{112}^{110} = \left(\frac{1}{3}\right)\epsilon_{110}^{110} + \left(\frac{2}{3}\right)\epsilon_{001}^{110}. \quad (7)$$

The superscripts denote the stress direction; the subscripts on ϵ , T denote the diagonal component of the tensor in the indicated direction. ($\epsilon_{110} \equiv \epsilon_{jj}$ with j in the 110 direction.)

We also write down the results for the stress in a (100) direction:

$$\epsilon_{100}^{100} = [W_{11}s_{11} + 2W_{12}s_{12}]T_{100}, \quad (8)$$

$$\epsilon_{001}^{100} = [W_{11}s_{12} + W_{12}(s_{11} + s_{12})]T_{100}. \quad (9)$$

The fundamental constants W_{11} , W_{12} , and W_{44} are plotted in Fig. 3. W_{44} was obtained using Eqs. (4) and (5) with the data of Fig. 2(a), while W_{11} and W_{12} were obtained from Eqs. (8) and (9) with the data of Fig. 2(d). The units for the W_{ij} are arbitrary but are the same for all three curves.

As symmetry checks we plot in Fig. 2(c) the measured ϵ_{112}^{110} with the value calculated from Eq. (7). A further symmetry check is obtained by comparing in Figs. 2(b) and 2(d) the measurements of ϵ_{001}^{100} and ϵ_{001}^{110} which Eqs. (9) and (6) show should be identical. The comparisons

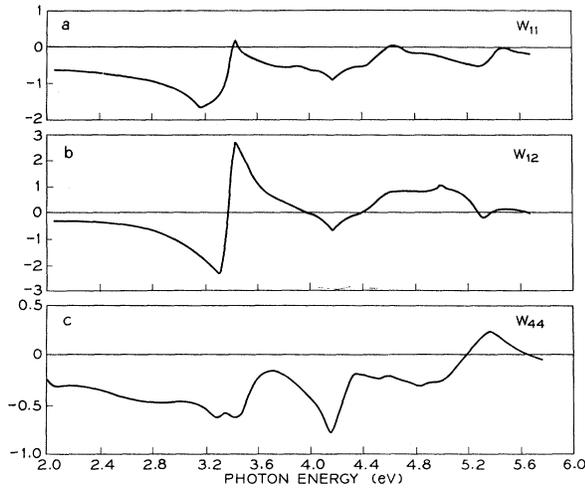


FIG. 3. Fundamental piezorefractance constants as functions of frequency.

are seen to be quite close.

In the vicinity of a critical point, there is a nonanalytic contribution to ϵ_2 which has the form

$$\epsilon_2 = a_g (\omega - \omega_c)^{1/2}, \quad \omega > \omega_c, \quad (10)$$

for type M_0 and M_2 critical points in the notation of Brust,⁸ or

$$\epsilon_2 = a_l (\omega_c - \omega)^{1/2}, \quad \omega < \omega_c, \quad (11)$$

for type M_1 and M_3 critical points. We ignore the analytic parts of ϵ_2 because, upon differentiation, only the nonanalytic term yields an infinity at ω_c . For the same reason, we ignore the strain dependence of a . Differentiation of Eq. (10) gives

$$\frac{d\epsilon_2}{du_{kl}} = b_g (\omega - \omega_c)^{-1/2}; \quad \omega > \omega_c, \quad (12)$$

$$b_g = \frac{-a_g}{2} \frac{d\omega_c}{du_{kl}}.$$

To obtain $d\epsilon_1/du_{kl}$ we differentiate the Kramers-Kronig relation and keep only the singular contribution, which is

$$\frac{d\epsilon_1}{du_{kl}} = \frac{b_g}{(\omega - \omega_c)^{1/2}}; \quad \omega < \omega_c. \quad (13)$$

The singular contributions from the derivatives of ϵ_1 and ϵ_2 are equal in magnitude but occur on opposite sides of the critical-point energy,

ω_c . Similarly, we find

$$\frac{d\epsilon_1}{du_{kl}} = \frac{-b_l}{(\omega - \omega_c)^{1/2}}; \quad \omega > \omega_c, \quad (14)$$

$$b_l = \frac{+a_l}{2} \frac{d\omega_c}{du_{kl}}.$$

The two cases are similar except for the reversal of sign.

Equation (2) shows that $d\epsilon_1/du_{kl}$ and $d\epsilon_2/du_{kl}$ have different weighting factors. This equation has been derived in scalar form by Seraphin,⁹ who has also computed the weighting factors from the n and k values of Philipp and Taft.¹⁰ At 3.3 eV the two coefficients are equal in magnitude but the range of equality is very narrow. The two peaks seen between 3.2 and 3.6 eV may then originate from the same critical point. Since the peaks are of opposite sign, the singularity must be of type M_1 or M_3 . The ϵ_2 data themselves require that it be type M_1 . At 4.2 eV the coefficient of $d\epsilon_2/du_{kl}$ goes through zero, hence only one peak is expected.

When the critical points lie on nondegenerate 100 or 111 symmetry axes, one can infer special symmetry relations between the singular contributions to the W coefficients due to symmetry relations among the deformation-potential constants of the critical points. A strain u_{xy} will not shift nondegenerate $\Delta(k00)$ energies, hence the contribution of Δ critical points to W_{44} will be zero. All nondegenerate Λ energies are shifted equally by a u_{xx} strain, hence the contribution of Λ critical points to W_{11} and W_{12} will be equal. Of course, both the Λ and Δ relations must apply to a nondegenerate Γ point.

Equality of W_{11} and W_{12} peaks is nowhere strongly suggested by the data. However, the strong peaks just above and below 3.4 eV in W_{11} and W_{12} are much weaker in W_{44} , which could be due to critical points at Δ . Brust's calculations¹¹ do indeed suggest that critical points on Δ make an important contribution to ϵ_2 at 3.4 eV. The valence state for the Δ critical points is doubly degenerate, however, so that the above conclusion would require that the sensitivity to u_{xy} strain of the degenerate valence bands be much less than the u_{xx} sensitivity of the nondegenerate conduction band. Gerhardt⁶ has also given evidence that the 3.40 peak comes from Δ critical points.

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MEASUREMENT OF THE MAGNON DISPERSION RELATION OF IRON*

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This note reports that the magnon dispersion curve for iron deviates considerably from the quadratic law, $\hbar\omega = Dq^2$, even for relatively small wave vectors q , indicating a long-range magnetic interaction in this metal.

The measurements were done by utilizing the diffraction method¹ in conjunction with polarized neutrons. The angular width of the magnon scattering surface was determined as a function of the angle $\Delta\varphi$ by which the crystal is misset from the elastic Bragg peak (110). (See Fig. 1.)

Correction for the instrumental resolution was made by folding the resolution function (measured by scanning the Bragg peak) into an ideal rectangular profile. A least-squares fit of the profile so obtained to the observed profile was made with the width as the varied parameter. The use of polarized neutrons in the present experiment allows one to distinguish unambiguously the magnon scattering from other scattering contributions, as has already been described in some detail.² The main part of the data were collected on a single crystal of iron containing 4 at.% silicon, kindly provided by Dr. H. J. Williams and Dr. A. J. Williams of Bell Telephone Laboratories.

The energy of a magnon with wave vector \vec{q} may be written as a function of the exchange interactions $J(\vec{r})$,

$$\hbar\omega = 2S \sum_{\vec{r}} J(\vec{r}) (1 - e^{i\vec{q} \cdot \vec{r}}), \quad (1)$$

assuming zero energy gap. For a body-centered lattice with \vec{q} along the [100] direction, this expression reduces to the following simple form if only the nearest neighbor interaction J_1 is effective:

$$\hbar\omega = 16J_1S[1 - \cos(\frac{1}{2}qa)]. \quad (2)$$

And for small q values,

$$\hbar\omega = 2J_1Sq^2a^2 \left(1 - \frac{q^2a^2}{24} + \frac{q^4a^4}{5760} + \dots \right). \quad (3)$$

Small-angle scattering measurements by Lowde and Umakantha and other workers at Harwell³ yield the result $\hbar\omega = Dq^2$, with $D = 286 \text{ meV } \text{\AA}^2$. This method corresponds to the special case of the diffraction technique with zero misset angle at the (000) reflection and effectively samples the dispersion curve for exceedingly small q values ($qa/2\pi < 0.025$), so that the higher terms in Eq. (3) are truly negligible.