Brillouin-Scattering Measurements on Silicon and Germanium

J. R. Sandercock
Laboratories RCA Ltd., Zürich, Switzerland
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This is a report of the first Brillouin scattering measurements on silicon and germanium. The measurements were made at $\lambda = 6328$ and 4880 Å at which frequencies the optical absorption lies in the range 10^4 to 6×10^5 cm^{-1}. As a result of the high absorption, the light sees only a few wavelengths' extent of the phonon, and so from the uncertainty principle the Brillouin peaks become very broad. The measured linewidths have been analyzed to give the real and imaginary part of the refractive indices. Good agreement is obtained with the values reported in the literature.

Up to the present time very few results have been reported of Brillouin scattering measurements on absorbing materials. The reason is that such measurements must be made in the backscattering configuration where, using the conventional Fabry-Perot interferometer, the large amount of Rayleigh-scattered light arising from surface defects completely swamps the much weaker Brillouin components. Recently, a new technique using a multipassed Fabry-Perot has been described[1,2] which, by increasing the contrast of the instrument, enables an improvement by a factor of 10^5 or more in the rejection of the Rayleigh component. By using this technique the measurement of Brillouin scattering in absorbing materials becomes straightforward.

Of particular interest in such measurements is the large width of the Brillouin components which is a direct manifestation of the high absorption. One can understand this in a simple manner as follows. Consider radiation of vacuum wave vector k_0 incident normally on a medium of refractive index $n = n_1 + in_2$. For light scattered through 180°, a transfer of momentum of $2n_1k_0\hbar$ must occur. If $n_2 \ll n_1$, a phonon of momentum $p = 2n_1k_0\hbar$ is involved and the corresponding frequency shift $\Omega_0 = 2n_1k_0v$, where v is the hypersound velocity. However, if $n_2 \simeq n_1$, the light intensity falls off rapidly inside the medium, falling to $1/e$ of the incident intensity within a distance $(2n_2k_0)^{-1}$. The phonon is thus observed within a distance of the order $(2n_2k_0)^{-1}$ from the surface, and so from the uncertainty principle the experiment measures the phonon momentum only to within $\Delta p \simeq 2n_2k_0h$ around the mean value $p = 2n_1k_0\hbar$. The Brillouin peak, therefore, has a width $\Delta\Omega \simeq 4\pi n_2k_0v$; the ratio of width to energy shift, $\Delta\Omega/\Omega_0$, is of the order of $2\pi n_2/n_1$. The exact relation is determined later to be $\Delta\Omega/\Omega_0 = 2n_2/n_1$.

The measurement of the Brillouin spectrum thus determines (a) the product n_1v from the shift and (b) the product n_2v from the linewidth, assuming n_2 to be sufficiently large that phonon lifetime effects do not contribute significantly to the linewidth.

Normally in measurements on transparent crystals the refractive index is known very precisely and the Brillouin shift is used to determine the hypersound velocity v. In the present measurements, however, the refractive indices are rather imprecisely known. By constrast the ultrasound velocities are accurately known[3] and it seems quite reasonable in this case to equate the hypersound velocity to the ultrasound values and use the Brillouin measurements to determine the optical constants n_1 and n_2.

First it is necessary to investigate more closely the case of light scattering from a nontransparent medium. Light scattering from transparent cubic crystals has been treated very lucidly by Benedek and Fritsch.[4] Their treatment may be extended quite generally to the case of nontransparent materials, but the calculation is particularly simple for the case of 180° backscattering. Since, in practice, backscattering experiments do not depart greatly from the 180° configuration, particularly for materials of high refractive index, we will indicate here the theory only for this simplified configuration.

By allowing for the absorption of both the incident and scattered radiation, one may show, following Ref. 4, that the backscattered field E_s at the point $\vec{R}$ is given by

$$E_s(R,t) \sim \exp[i(\vec{k}\cdot\vec{R} - \omega t)] \int d^3r \int d^3q\, \delta\alpha(\vec{q}) \exp\{i[n(\vec{k}_0 - \vec{k}) - \vec{q}]\cdot\vec{r}\} \quad (1)$$

with $|k| = |k_0| \pm \Omega(q)/c$ and $\omega = \omega_0 \pm \Omega(q)$, where $\vec{k}$, ω refer to the scattered radiation, $\vec{k}_0$, ω_0 to the incident

radiation, and $\vec{q}$, $\Omega(q)$ to the phonon; R and r are measured from the surface of the medium. The fluctuation $\delta\alpha(q)$ in the polarizability is related through the Pockels coefficients to a strain $e(\vec{q})$. Provided that $\hbar\Omega(q) \ll k_B T$, we have $e^2(\vec{q}) \sim k_B T$ and so the fluctuation $\delta\alpha(\vec{q})$ may be taken as independent of $\vec{q}$. If we now define $\hat{z}$ as the direction of k_0 and ignore the small difference between $|k_0|$ and $|k|$, we obtain from (1)

$$E_s(R, t) \sim \delta(\vec{q} \times \vec{z}) \int \frac{dq_z}{i(2nk_0 - q_z)}.$$

This result differs from the normal expression for E_s in backscattering, viz.,

$$E_s \sim \delta(2n_1 k_0 - q_z),$$

because of the finite value of n_2.

The power P_s scattered per unit solid angle around the direction R is proportional to $|E_s(R, t)|^2$. Since the contributions from the different phonons q_z are independent, we may write

$$P_s(R, t) \sim \int \frac{dq_z}{(2n_1 k_0 - q_z)^2 + (2n_2 k_0)^2} \sim \int \frac{d\Omega}{(\Omega_0 - \Omega)^2 + [(n_2/n_1)\Omega_0]^2},$$

where the phonon frequency $\Omega_0 = 2n_1 k_0 v$. The power $(dP_s/d\Omega)(\omega_0 \pm \Omega)d\Omega$ in the frequency range $d\Omega$ is thus given by

$$\frac{dP_s}{d\Omega}(\omega_0 \pm \Omega)\, d\Omega \sim \frac{d\Omega}{(\Omega_0 - \Omega)^2 + [(n_2/n_1)\Omega_0]^2}. \qquad (2)$$

The Brillouin scattered light thus has a Lorentzian distribution centered about $\omega = \omega_0 \pm \Omega_0$ with a full width at half-maximum of $\Delta\Omega = (2n_2/n_1)\Omega_0$, or expressed as a relative width,

$$\Delta\Omega/\Omega_0 = 2n_2/n_1.$$

Backscattering measurements on germanium and silicon have been made which essentially confirm these results. For convenience we used a scattering angle of 90° outside the crystal. Since the refractive indices of silicon and germanium are high (around 5 in the visible region) both the incident and scattered radiation was traveling nearly perpendicularly to the surface, and so the experiment measured nearly 180° backscattering. Most of the measurements were made with the incident light polarized in the scattering plane. For these measurements a useful gain (×2) in scattered intensity was obtained by making the incident light strike the crystal at the Brewster angle. Furthermore the scattered light was then more nearly at normal incidence thus eliminating relative intensity variations due to different reflection losses for the differently polarized scattered components.

The measurements were made at $\lambda = 6328$ Å (20 mW) and $\lambda = 4880$ Å (200 mW), the scattered light being detected by using the multipassed interferometer previously described.[1,2] We used broad-band dielectric mirrors having about 93% reflectivity. The RCA C 31000 F photomultiplier was cooled and magnetically defocused to give a dark count rate of about $\frac{1}{2}$/sec. Despite the high contrast of the interferometer, which exceeds 10^9, the surface quality of the crystals had to be good. Measurements were performed on (100) and (111) surfaces of pure germanium and silicon. While the commercially obtained silicon surfaces were excellent, some trouble was had in obtaining good germanium surfaces. The best results were obtained from CP4-etched surfaces, but the Rayleigh scattered light was still so intense as to cause heating of the photomultiplier cathode. As a result the interferometer scan rate had to be reduced to about 10/min in order to avoid a tail of dark counts in the measured spectra.

The crystals were held at room temperature but some heating of the scattering volumes was inevitable. While it was not possible to measure this, it is estimated that this heating should not have exceeded about 30°K.

Some typical spectra are shown in Fig. 1. For both silicon and germanium, measurements on (111) faces gave longitudinal and doubly degenerate transverse phonon peaks of roughly equal peak intensity. In contrast, for measurements on the (100) faces the transverse phonon peak was virtually absent. The three spectra of Fig. 1 show clearly the effect of absorption on the Brillouin peak width. In 1(a) where the absorption coefficient is about 10^4 cm^{-1}, the peaks are sharp, having a width-to-shift ratio of about 3%. At the other end of the scale is 1(c) showing the corresponding spectrum for germanium at the same wavelength where the absorption is about 6×10^5 cm^{-1}. The width-to-shift ratio is about 100% here. At an intermediate absorption spectra 1(b)

demonstrates clearly for the transverse and longitudinal phonons the increase in linewidth with increasing energy shift.

Lorentzians were fitted to all the measured spectra with the exception of the one measurement on silicon at $\lambda = 6328$ Å where the rather weak spectrum allowed only the energy shift to be measured. The Lorentzians were corrected for the instrumental profile (the finesse was about 55) and for the finite number of sampled points in the recorded spectrum. From Eq. (2) the products $n_1 v$ and $n_2 v$ were then obtained, where v is the appropriate hypersound velocity. Taking the values of v as equal to the known ultrasound velocities,[3] the values of n_1 and n_2 were calculated (see Table I). For comparison various values have been taken from the literature. For silicon the most reliable value of n_1 for the bulk material is probably that of Vedam, Knausenberger, and Lukes[5] whose measurements, however, were restricted to $\lambda = 5461$ Å. The values of n_1 quoted in Table I are an extrapolation of this measurement using the spectral dependence reported by Philipp and Taft.[6] The value for silicon of n_2 from transmission measurements on thin single crystals[7] is considered by the authors to be reliable only to within 20%. For germanium the results of reflection measurements by Potter[8] and Archer[9] are the best available, but there is considerable disagreement between the two. The transmission measurements of Harbeke[10] differ again. Other reported results involving a Kramers-Kronig analysis of measurements without allowance for surface films are not reliable.

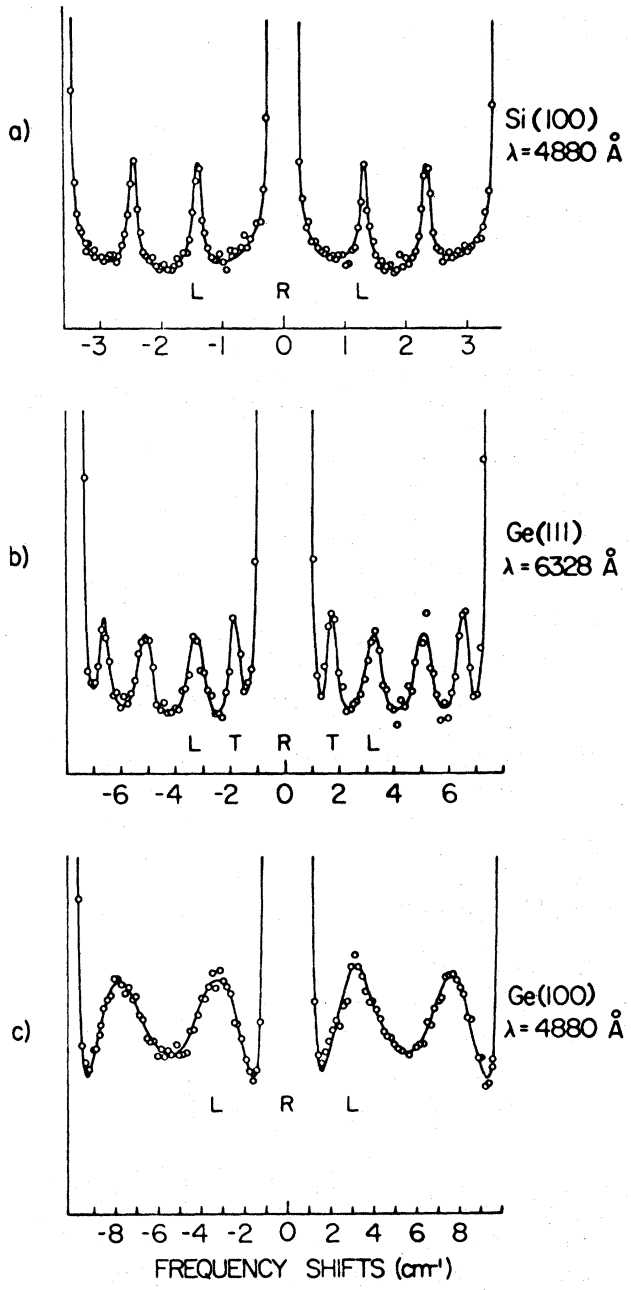

FIG. 1. Some typical spectra, showing absorption-broadened Brillouin peaks. In (a) the peaks are observed in the first neighboring order, in (b) and (c) in the zero order. The recording times were about 2, 4, and 10 h, respectively.

From Table I it is seen that the Brillouin measurements on silicon yield values of n_1 in excellent agreement with the reflectivity measurements while the less precise value of n_2 is reasonably close to the transmission measurement value. For germanium, of the reported values, the measurements of Potter[8] agree most closely with the Brillouin results. In view of the very large spread in the reported values[8-10] the agreement is seen to be good. It is interesting that the greatest difference between the present and reported values occurs in the most highly absorbing measurements where the reported values differ most strongly among themselves. It should be emphasized the Brillouin measurements determine the optical constants of bulk material, and are unaffected by the presence of surface films.

To conclude, a multipassed interferometer has been used to obtain Brillouin measurements on silicon and germanium in the highly absorbing range between 10^4 and 6×10^5 cm^{-1}. It has been shown that as a result of the finite value of n_2, the uncertainty principle prevents the geometry of the experiment from determining uniquely the phonon wave vector. Since the Brillouin energy shift is proportional to the phonon wave vector,

TABLE I. Comparison of the measured optical constants with values taken from the literature.

λ (Å)	Direction and mode	n_1 Present	n_1 Previous	n_2 Present	n_2 Previous
		Silicon			
6328	[100], L	3.89	3.92 [a]	...	...
	[100], T	3.92		...	...
4880	[100], L	4.38	4.36 [a]	0.06	0.051 [b]
	[111], L	4.35		0.07	
		Germanium			
6328	[100], L	5.56		0.67	
	[111], L	5.61	5.53,[c] 5.43[d]	0.70	0.69,[c] 0.82,[d] 0.85[e]
	[111], T	5.55		0.65	
4880	[100], L	5.76	4.55,[c] 4.31[d]	2.38	2.56,[c] 2.30,[d] 1.55[e]

[a]Refs. 5, 6. [c]Ref. 8. [e]Ref. 10.
[b]Ref. 7. [d]Ref. 9

the peaks are absorption broadened, as distinct from Raman scattering where the optical phonon energy is virtually independent of wave vector. Analysis of the Brillouin spectra allows the measurement of both the optical constants (assuming that the hypersound velocity is known). These Brillouin results have been shown to agree closely with values reported in the literature, and are probably at least as reliable in this high absorption region. The multipassed interferometer is seen to be a powerful tool for the investigation of Brillouin scattering in opaque materials.

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Critical Exponents in 3.99 Dimensions*

Kenneth G. Wilson and Michael E. Fisher
Laboratory of Nuclear Studies and Baker Laboratory, Cornell University, Ithaca, New York 14850
(Received 11 October 1971)

Critical exponents are calculated for dimension $d = 4 - \epsilon$ with ϵ small, using renormalization-group techniques. To order ϵ the exponent γ is $1 + \frac{1}{6}\epsilon$ for an Ising-like model and $1 + \frac{1}{5}\epsilon$ for an XY model.

A generalized Ising model is solved here for dimension $d = 4 - \epsilon$ with ϵ small. Critical exponents[1] are obtained to order ϵ or ϵ^2. For $d > 4$ the exponents are mean-field exponents[1] independent of ϵ; below $d = 4$ the exponents vary continuously with ϵ. For example, the susceptibility exponent γ is $1 + \frac{1}{6}\epsilon$ to order ϵ for $\epsilon > 0$, and 1 exactly for $\epsilon < 0$. The definitions for nonintegral d are trivial for the calculations reported here but may be more difficult for exact calculations to higher orders in ϵ. The exponents will be calculated using a recursion formula derived elsewhere[2] which represents critical behavior approximately in three dimensions but turns out to be exact to order ϵ (see the end of this paper). Exponents will also be obtained for the classical planar Heisenberg model (XY model) and a modified form of Baxter's eight-vertex model.[3]