ly along with the results of other experiments.

We are indebted to Mr. J. Cozzo for his expert technical assistance. We wish to thank Professor Joffrin and Professor Levelut for informing us of their results prior to publication and for supplying the sample of SbSI.

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Low-Temperature Thermal Brillouin Scattering in Fused Silica and Borosilicate Glass*

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Brillouin scattering experiments have been performed between 1.7 and 300 K in glasses to demonstrate the existence of thermal phonons. The theoretically expected linear temperature dependence of the Brillouin peak intensities has been observed.

In crystalline solids, the existence of Debye thermal lattice excitations or phonons can be demonstrated convincingly through specific-heat measurements. It is well established that the low-temperature lattice specific heat of pure crystals agrees to within percents with that calculated on the basis of the Debye model, using experimental sound velocities. In noncrystalline dielectric solids, however, it has recently been shown that the low-temperature specific heat C_v does not obey the Debye law.[1,2] Instead, it was found that C_v can be expressed as a polynomial,

$$C_v = c_1 T + c_3 T^3, \tag{1}$$

where c_3 is up to twice as large as predicted by the Debye model. This observation has given rise to the fundamental question whether Debye-like plane-wave thermal excitations exist at all in glasses at low temperatures, i.e., whether one is justified in writing

$$C_v = c_1 T + (c_3' + c_{\text{Debye}}) T^3, \tag{2}$$

where both $c_1 T$ and $c_3' T^3$ are caused by the dis-

order. The question had arisen in connection with a study of the thermal conductivity in glasses, in which the possibility had been discussed that the heat was not carried by Debye phonons.[1,2] As an alternative, it had been suggested that the phonon lifetimes could be so short that a separation of c_3 as written in Eq. (2) would not be justified. Recent low-temperature (0.1 to 1 K) ultrasonic attenuation measurements in the frequency range of 10^9 sec^{-1} have not been able to answer this question.[3,4] Rather, it was found that the attenuation increased with decreasing pulse intensity, demonstrating strong nonlinear forces in the glassy solid. From these measurements, an extrapolation to ultrasonic intensities characteristic for thermal phonons was not possible.

In this Letter, we want to present experimental evidence that thermal phonons in the frequency range of 2×10^{10} sec^{-1} do indeed exist in glasses in the temperature range 1.7 to 300 K, and that their lifetimes are at least 10 vibrational periods. Earlier spontaneous Brillouin scattering experiments in glasses were done at temperatures

above 80 K[5,6]; because of the weak intensity of the Brillouin scattered light at low temperatures, it was necessary for the present experiment to employ an intense incident light source along with high-sensitivity photon-counting detection.

We used a Coherent Radiation, model 52, argon-ion laser, operating in a single longitudinal mode, at 5145 Å, delivering 300 mW of linearly polarized light. The scattered light was collected at an angle of 90° with $f/4$ optics. An I_2 vapor cell ($T \sim 60°C$) was used to filter out most of the unshifted (Rayleigh) laser light.[7] An interference filter, 30-Å band pass, eliminated fluorescent light from the sample. The Fabry-Perot was pressure scanned; its plates were dielectric coated for 98% reflectivity at 5145 Å and flat to $\lambda/100$. One scan (50 GHz) took 9 min. The photoelectric pulses from an EMI 6256S photomultiplier were processed with photon counting elec-

tronics, discriminating against small pulse heights.

Figure 1 shows the Brillouin spectra at 230 and 1.7 K in fused silica. The Stokes peaks for one order are shown with the anti-Stokes peaks for the next higher order. At low temperature the transverse phonon peaks are smaller than the noise. The longitudinal anti-Stokes peak at 230 K appears broader than the Rayleigh peaks because of its overlap with the Stokes peak. Spectra were also taken for borosilicate glass (Corning 7740). In this case, the longitudinal peaks have smaller frequency shifts and the Brillouin linewidths are equal to the Rayleigh widths to within 10% at all temperatures. The temperature dependence of the Brillouin peak intensities for those peaks which are unaffected by the I_2 filter is shown in Fig. 2. The data were taken as the Dewar slowly (~ 6 h) warmed up to room temper-
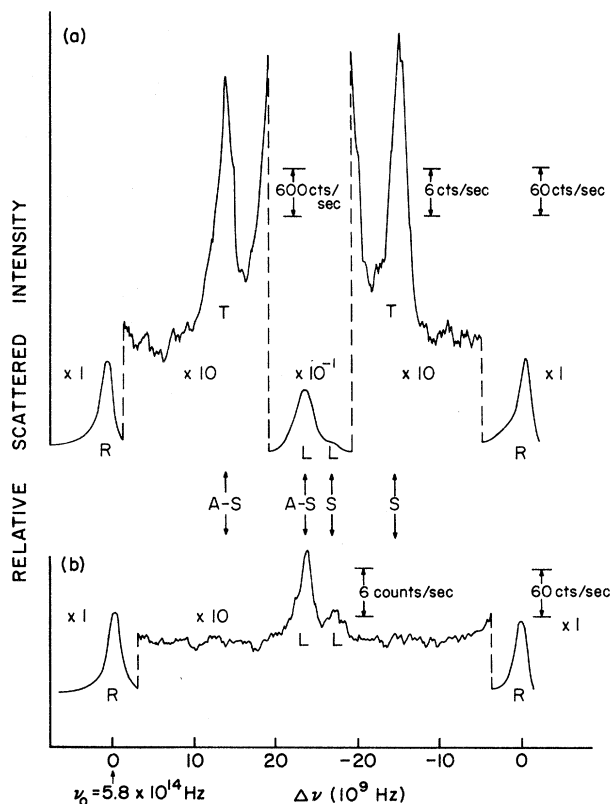

FIG. 1. Brillouin spectra of fused silica at (a) 230 K, (b) 1.7 K. Rayleigh (R), longitudinal (L), and transverse (T) peaks are shown for the anti-Stokes (A-S) and Stokes (S) shifts. Gain changes, as noted relative to the Rayleigh peak, were made as necessary (at dashed lines). The longitudinal Stokes peak is cut by the I_2 filter. The Rayleigh peaks in each scan belong to adjacent orders.
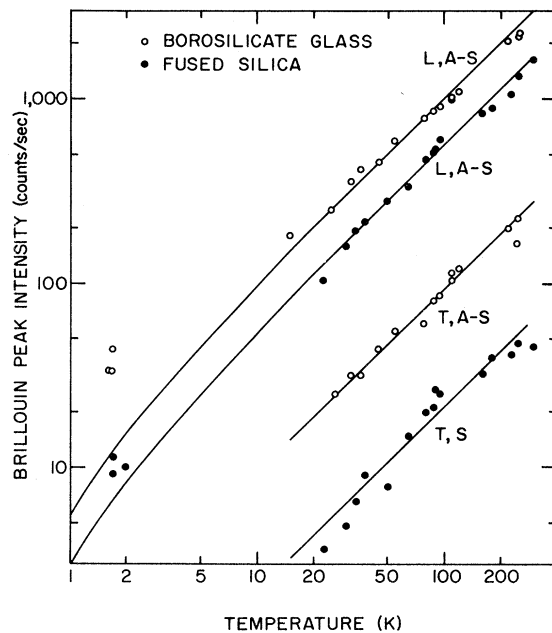

FIG. 2. Brillouin peak intensities for borosilicate glass (open circles) and for fused silica (closed circles) as a function of temperature. The theoretically expected temperature dependence of the intensities is indicated by the solid curves. The anti-Stokes (A-S) intensity is proportional to the phonon occupation number $\langle n \rangle$, where $\langle n \rangle = [\exp (h|\Delta\nu|/k_B T) - 1]^{-1}$, while the Stokes (S) intensity is proportional to $\langle n \rangle + 1$. For $T \gg h|\Delta\nu|/k_B$ (i.e., $T \gg 1$ K), both A-S and S intensities vary linearly with temperature. For $T \lesssim h|\Delta\nu|/k_B$, the A-S intensity should approach zero exponentially, while the S intensity should become temperature independent. "L" and "T" represent longitudinal and transverse peaks, respectively.

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ature. Data could not be taken between 2 and 20 K because of the rapid initial warmup. The 1.7-K data were taken with the sample immersed in liquid helium. As expected theoretically for spontaneous Brillouin scattering, the intensities vary linearly with temperature for $T \gg h|\Delta\nu|/k_B$. The longitudinal anti-Stokes peaks at 1.7 K are higher than expected and were probably caused by sample heating by the laser. This heating was larger in the Corning 7740 glass; it also fluoresced more strongly than the fused silica. In some of the 1.7-K spectra of borosilicate glass there appeared the hint of a transverse peak above the noise. The peak intensity was larger than theoretically expected and is consistent with sample heating. Its width was equal to the Rayleigh width.

Table I summarizes the results. The hypersonic sound velocity is found from the familiar relation $\Delta\nu = \pm 2\nu_0 n(v/c)\sin\frac{1}{2}\theta$, where $\Delta\nu$ is the frequency shift of the Brillouin line, ν_0 the incident light frequency, n the refractive index, v the sound velocity, c the speed of light in vacuum, and θ the scattering angle (here 90°). In calculating v from the experimental frequency shifts the very small temperature variation of the refractive index was neglected, and the room temperature values were used. Phonon lifetime effects would broaden the Brillouin line over that caused by the instrumental width, by the amount $\delta\nu$ (measured full width at half-height). The amplitude attenuation coefficient α is related to this additional Brillouin linewidth $\delta\nu$ by $\alpha = \pi \, \delta\nu/v$. From our linewidth data we estimate that the full width at half-height of the Brillouin peak is less than 20% larger than the Rayleigh width, hence $\delta\nu < 0.2 \times (2.1\text{ GHz}) = 420$ MHz at all temperatures for both glasses. This gives upper bounds on α as noted in Table I. From these values follow lower limits for the phonon mean free path (for energy decay) using $\bar{l} = 1/2\alpha$. Previous determinations of the phonon mean free path had been based on measurements of the thermal conductivity κ of glasses.[1] Under the *assumption* that the heat was carried entirely by Debye plane-wave phonons, $\bar{l}$ was computed from κ and the measured Debye sound velocity v_{Debye} using the gas kinetic formula $\kappa = \frac{1}{3}c_{\text{Debye}}v_{\text{Debye}}\bar{l}$. As shown in Table I, the lower limits of $\bar{l}$ determined from Brillouin scattering are smaller than the average mean free paths derived from κ. Thermal Brillouin scattering experiments, to measure lifetime broadening in the liquid-helium temperature range for these glasses, would require a factor-of-10 higher resolution to observe a broadening ~ 20% larger than the instrumental width, assuming that both longitudinal and transverse phonons are equally damped. Such measurements are not possible with the present equipment.

These measurements show that thermal pho-

TABLE I. Hypersonic velocity and attenuation obtained from the present measurements. In the last two columns, the phonon mean free path obtained from the Brillouin scattering is compared with that obtained from thermal conductivity measurements, Refs. 1 and 2. Note that these mean free paths $\bar{l}$ are connected to the hypersonic amplitude attenuation coefficient α by $\bar{l} = 1/2\alpha$, since $\bar{l}$ measures energy attenuation.

Material	Brillouin Component	Brillouin Shift (GHz)	Refractive Index at 300 K	Hypersonic Velocity (10^5 cm/sec)	Phonon Wavelength (Å)	Amplitude Attenuation Coeff. α (cm^{-1})	Phonon Mean Free Path $\bar{l}$ (cm) from $\frac{1}{2\alpha}$ this work	Phonon Mean Free Path $\bar{l}$ (cm) from thermal conductivity[e]
Fused Silica[a]	L	23.5	1.462[c]	$5.85 \pm .10$	2490	$< 2.0 \times 10^3$	$> 2.5 \times 10^{-4}$	4×10^{-3}
	T	14.8		$3.68 \pm .10$	2490	$< 3.2 \times 10^3$	$> 1.6 \times 10^{-4}$	
Borosilicate Glass[b]	L	22.5	1.478[d]	$5.53 \pm .07$	2460	$< 2.4 \times 10^3$	$> 2.1 \times 10^{-4}$	2×10^{-3}
	T	14.0		$3.45 \pm .05$	2460	$< 3.8 \times 10^3$	$> 1.3 \times 10^{-4}$	

[a]Suprasil I; manufacturer, Quarzschmelze Heraeus, Germany.

[b]Corning No. 7740; approximate composition (by weight): SiO_2 80.5%, B_2O_3 12.9%, Na_2O 3.8%, Al_2O_3 2.2%, K_2O 0.4%, and Li_2O 0.2%.

[c]American Institute of Physics Handbook (McGraw-Hill, New York, 1972).

[d]C. Parker, Corning Glass Works, private communication.

[e]The mean free path is obtained from the thermal conductivity at that temperature for which the phonons have a dominant wavelength of 2500 Å (0.3 K). See Fig. 9 of Ref. 1 (dominant phonon approximation).

nons exist as good normal modes in glasses at
temperatures as low as 1.7 K. We conclude that
the experimental specific heat must indeed be
written in the form of Eq. (2), i.e., the Debye
specific heat is simply masked by the specific
heat caused by the disorder. From the lower lim-
its of the phonon lifetimes determined in our ex-
periment it follows that thermally excited Debye
phonons must contribute to the heat transport in
glasses at low temperatures. The questions
whether the additional excitations observed in
specific heat also carry heat, or whether they
only scatter the Debye phonons, or, finally,
whether the additional excitations are involved
at all in the heat flow, require further studies.

I am indebted to R. O. Pohl for stimulating this
work and for assistance in the preparation of the
manuscript. I wish to thank him, P. L. Hartman,
R. H. Silsbee, J. Vitko, and R. B. Stephens for

encouragement and helpful discussions.

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Diffusivity of Charge Carriers in Semiconductors in Strong Electric Fields*

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(Received 30 July 1973)

Similarities between diffusion of hot charge carriers in semiconductors and in gases
are discussed, and formulas for $D_{\parallel}$ and $D_{\perp}$, the diffusion coefficients parallel and per-
pendicular to the applied electric field, are given. It is shown that anisotropy of diffu-
sion can be strong even when the carrier distribution function is nearly isotropic in mo-
mentum space.

Development of techniques[1,2] capable of accu-
rate measurement of the diffusivity of charge
carriers in a semiconductor parallel and perpen-
dicular to a strong electric field $\vec{E}$ opens the way
for a more thorough understanding of the nature
of the interaction between carriers and lattice.
The parallel and perpendicular diffusion coeffi-
cients, $D_{\parallel}$ and $D_{\perp}$, respectively, have quite a
different qualitative dependence[1-3] upon the field
from that of the mobility coefficient μ, and may
therefore yield information not obtainable from
mobility data alone. Unfortunately, a satisfac-
tory quantitative theory of hot-carrier diffusion
in semiconductors is not available, and so the
link between diffusion experiments and charge-
carrier–phonon interaction cannot be precisely
established. Even qualitative aspects are not
properly understood: It is a common misconcep-
tion that the anisotropy of diffusion in an electric
field (i.e., $D_{\parallel} \neq D_{\perp}$) arises *solely* from the asym-

metry (elongation) of the carrier distribution func-
tion, $f(\vec{k})$, in momentum space.[3,4] Further, it
has not been recognized that diffusion of charge
carriers in a semiconductor is in many ways
analogous to diffusion of electrons or ions in a
gas of neutral atoms. The theory of the latter
problem has received a great deal of attention
recently,[5-7] and we can use some of these results
to provide at least a starting point for a theory
appropriate to semiconductors.

In relation to ions diffusing in a gas, Wannier[8]
conjectured the following generalization of the
Einstein relation for diffusion parallel to $\vec{E}$:

$$D_{\parallel} = \frac{\kappa T_{\parallel}}{e}\mu\left(1 + \frac{\partial \ln\mu}{\partial \ln E}\right), \tag{1}$$

a formula recently substantiated via nonequilib-
rium thermodynamics.[9] It was also shown in Ref.
9 that the perpendicular diffusion coefficient is