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Brillouin scattering in vitreous silica below 1 K

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We report the first measurements of Brillouin frequency shifts and linewidths in a solid below 1 K. The resonant attenuation of 35-GHz phonons by two-level systems in vitreous silica and the related anomalous behavior of the velocity are observed down to 0.3 K, where the phonon energy $\hbar\omega \simeq 5kT$. The low-temperature data are fully described by current theory based on the existence of two-level systems in glasses. Above 10 K, in contrast, a qualitative agreement between theory and experiment is found.

I. INTRODUCTION

At low temperatures, amorphous materials exhibit anomalous properties[1] which are attributed to the existence of two-level systems (2LS). These 2LS have a nearly constant density of states of their energy and interact strongly with phonons resulting in unusual elastic properties.[2] Since their discovery[3] many acoustic experiments have been carried out at low temperatures. Conventional ultrasonic experiments, however, become very difficult at frequencies above 1 GHz because of the strongly increasing absorption. As a consequence, the investigation of the interaction of 2LS with ultrasonic phonons in the interesting regime $\hbar\omega > kT$, i.e., where most of the interacting 2LS are in their ground state, had to be carried out at ultralow temperatures.[4]

A drastic increase in frequency range, however, can be achieved by probing optically the elastic properties of solids using Brillouin scattering (BS). Here the velocity and attenuation of hypersonic waves is deduced from the position and linewidth of the Brillouin line, respectively. With this method, even high absorptions where the mean free path of phonons extends over several wavelengths only can be easily observed. Although Brillouin scattering has already been applied to study the elastic properties of glasses down to temperatures of 2 K,[5] the limit where $\hbar\omega > kT$ has not been attained in these experiments. In this paper we report the first BS experiments in a solid well below 1 K. We have observed the velocity and absorption of 35-GHz phonons in vitreous silica down to temperatures of 0.3 K. We find that the acoustic anomalies of glasses, which have been observed ultrasonically at low frequencies, are still characteristic for the behavior of 35-GHz phonons.

II. EXPERIMENTAL

One of the major problems in the study of Brillouin spectra is that the weak Brillouin lines have to be measured on the tail of a strong Rayleigh line due to reflection or elastic scattering of the laser light by the end surfaces, defects, or frozen-in fluctuations in the sample. The contrast, defined as the ratio of the maximum to the minimum transmission of a given monochromatic line by the spectrometer, has then to be high enough to allow the separation of the Brillouin lines. On the other hand, the width of the Brillouin lines is only a few MHz at low temperatures. A high-resolving power $\nu/\Delta\nu$, where ν is the frequency and $\Delta\nu$ the bandwidth of the spectrometer, is needed for the analysis of these profiles.

The experiment was carried out using an arrangement of a triple-pass plane and a confocal Fabry-Pérot interferometers in tandem. The triple-pass Fabry-Pérot was used as a filter, in order to increase the contrast of the spectrometer. This interferometer was not scanned, and its maximum transmission was adjusted on the frequency of the Brillouin line under investigation. The confocal Fabry-Pérot was piezoelectrically scanned in order to analyze the spectra. In this way a high contrast (better than 10^{10}) and a high-frequency resolution ($\simeq 10^8$), corresponding to a bandwidth of about 5 MHz are combined with a high luminosity and sensitivity. Experimental details are described in Ref. 6. The experiments were performed in near backscattering geometry, which offers the following advantages: (i) the maximum frequency attainable in Brillouin scattering is reached (35 GHz in vitreous silica); (ii) the uncertainty of velocity measurements due to imperfect knowledge of the scattering angle θ is reduced to a minimum (changing the scattering angle by 10′ leads to a relative variation of 20 ppm of the velocity when $\theta = 177°$); (iii) the parasitic broadening of the line due to the finite aperture becomes negligible (less than 2 MHz in our experiments).

The intensity of the Stokes and anti-Stokes Brillouin lines is proportional to $\langle n\rangle + 1$ and $\langle n\rangle$, respectively, where $\langle n\rangle$ is the average number of thermally excited phonons. Therefore the signal detected at the peak of the Brillouin line dropped from 10 000 counts per second at room temperature to only 50 counts per second at 2 K. Below this temperature, the intensity of the Stokes line remains nearly constant. However, the intensity of the laser had to be strongly reduced in order to avoid sample heating and excessive ^{3}He vaporization. Finally, we detected 0.3 cps at 0.35 K (the dark counting rate was 0.1 cps). Typical measuring times were 1 h at 1 K and 5 h at the lowest temperature. The accuracy of the velocity and attenuation measurements of course depends on the signal-to-noise ratio and therefore decreases with decreasing temperature. However, using an original technique which has been described elsewhere,[7] we were able to measure the frequency shift with a precision of 1 MHz. The accuracy of the linewidth measurements was about 2 MHz.

The experiments were performed using a ^{3}He cryostat with antireflection coated optical windows. The sample was enclosed in a chamber filled with ^{3}He gas, and directly mounted at the bottom of the liquid ^{3}He copper chamber. Therefore, cooling was achieved both by conduction and by convection of the exchange gas. The temperature was measured at the copper plate close to the sample by a germanium resistor. The lowest temperature of 0.35 K was attained by using a charcoal adsorption pump. At this temperature, the cryogenic power was high enough to allow operation for about 6 h, with the incident laser power reduced to 10 mW in order to reduce the ^{3}He evaporation. This low power was used for all the measurements below 1 K. Experimentally, it was quite obvious, however, that this evaporation was caused by reflected and stray light absorbed by the ^{3}He copper chamber and not by the sample itself. The direct laser heating of the sample was tested at two temperatures (1.9 and 4 K): When the laser power was varied from 100 to 500 mW, the results for each temperature were identical within the accuracy of the experiments. Furthermore, in order to verify the temperature inside the scattering volume, we recorded simultaneously the Stokes and anti-Stokes lines and deduced its values from the well-known temperature dependence of their intensity ratio. Typical traces of spectra recorded at 1.9 and 0.45 K are shown in Figs. 1(a) and 1(b), respectively. At 1.9 K the intensity of the anti-Stokes line is 0.43 times that of the Stokes line, indicating a temperature of 1.99 K. The measured ratio of 0.05 at 0.45 K indicates a temperature of 0.55 K. From this analysis, we conclude that the temperature in the scattering volume is known with an accuracy of 0.1 K.

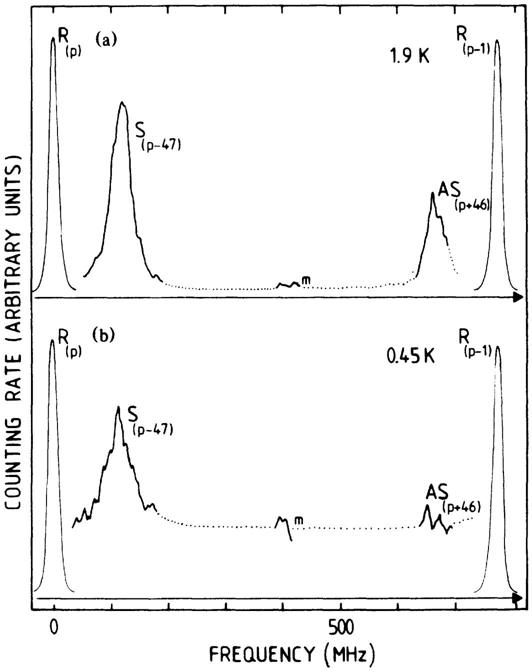

FIG. 1. Examples of spectra recorded at 1.9 K (a) and 0.45 K (b). S and AS are the Brillouin Stokes and anti-Stokes lines, respectively. The subscripts in brackets indicate the interference orders. R is a line at the laser frequency introduced artificially for defining the free spectral range and the instrumental profile. In order to increase the efficiency, each line as well as the minimum (m) of the transmitted function were recorded separately. Dotted line is a guide for eye only.

III. RESULTS

In Fig. 2 the acoustic absorption in vitreous silica Suprasil I is shown as a function of temperature. As

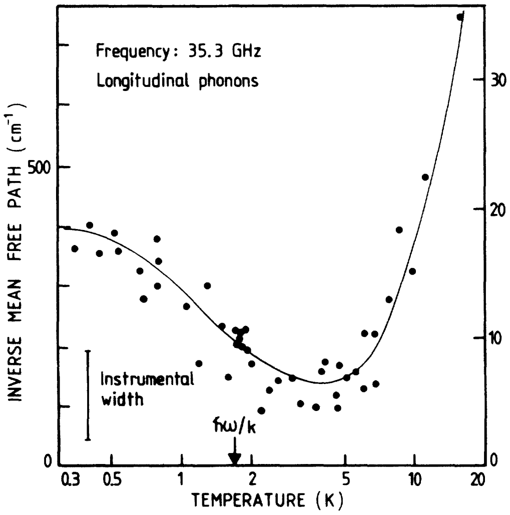

FIG. 2. Inverse mean free path as a function of temperature in vitreous silica. Solid line: best fit from Eqs. (1) and (3) (including a residual absorption of 50 cm^{-1} which corresponds to a 2.5-MHz linewidth).

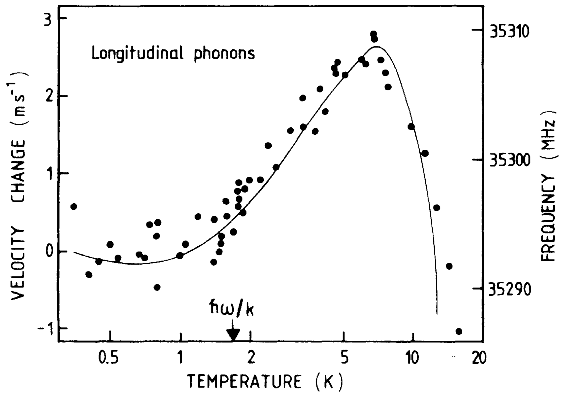

FIG. 3. Sound velocity as a function of temperature in vitreous silica. Solid line: best fit from Eqs. (2) and (3).

reported previously[5] the attenuation decreases on cooling to 4 K where the observed linewidth becomes comparable with the instrumental width of 7 MHz. At still lower temperatures the linewidth increases again and tends to a constant value at the lowest temperature of 0.35 K where $\hbar\omega \simeq 5kT$. In Fig. 3 the sound velocity is plotted as a function of temperature. It passes through a maximum around 7 K and decreases approximately logarithmically with temperature below the maximum.[5,8] Again at the lowest temperatures the velocity becomes temperature independent within the accuracy of our measurement.

IV. DISCUSSION

As mentioned the acoustic properties of glasses at low temperatures are mainly determined by 2LS. Below helium temperature the 2LS interact with phonons via the one-phonon process only: A phonon of energy $\hbar\omega$ is resonantly absorbed if its energy coincides with the energy splitting E of the interacting 2LS, i.e., if $E=\hbar\omega$. For this process a phonon mean-free-path l_{res} is calculated[2]:

$$l_{\mathrm{res}}^{-1}=\frac{\pi n(E)M^2E}{\hbar\rho v^3}\tanh\frac{E}{2kT} \quad . \tag{1}$$

Here v is the sound velocity, ρ the mass density, $n(E)$ the density of states, and M the deformation potential connected with the transition between the two states. In our further consideration we assume M to be constant, whereas $n(E)$ is only approximately constant for small energy values ($E \simeq 1$ K). Equation (1) is only valid in the limit of very small acoustic intensities. Since only thermally excited phonons are investigated, this condition is always fulfilled in our experiment.

This absorption process also causes a variation of the velocity of sound with temperature which can be calculated via Kramers-Kronig relation. If $n(E)$ and M are constant, an analytic expression can be found.[2] For $E=\hbar\omega \ll kT$ a well-known expression[2,8] results

$$\frac{\Delta v}{v}=\frac{n_0M^2}{\rho v^2}\ln\frac{T}{T_0} \quad . \tag{2}$$

Here $\Delta v=v(T)-v(T_0)$, where T_0 is an arbitrary reference temperature, which is 0.5 K in our considerations. In general the calculation has to be carried out numerically.

At higher temperatures ($T>1$ K) an additional absorption process becomes important: A sound wave traveling through the ensemble of 2LS modulates their level splitting. The 2LS try to relax into the new, instantaneous equilibrium via the exchange of phonons with the thermal bath. For this process the absorption and the related change in sound velocity are given by[2]

$$\left.\begin{matrix}-\Delta v/v\\ l_{\mathrm{rel}}^{-1}\end{matrix}\right\}=\frac{D^2}{2\rho v^2}\int_{E=0}^{\infty}\frac{\partial f}{\partial E}\,\frac{n(E)(2\omega^2\tau/v)^{\nu}}{1+\omega^2\tau^2}\,dE \quad , \tag{3}$$

where $f=[1+\exp(E/kT)]^{-1}$ and D is the deformation potential, determined by the shift in the level splitting for unit strain. The exponent $\nu=0$ is valid for the variation of the sound velocity and $\nu=1$ for the absorption. The relaxation time due to the one-phonon interaction is given by[2]

$$\tau^{-1}=\left(\frac{M_l^2}{v_l^5}+\frac{2M_t^2}{v_t^5}\right)\frac{E^3}{2\pi\rho\hbar^4}\coth\frac{E}{2kT} \quad . \tag{4}$$

The integration has to be carried out over all 2LS since all thermally excited systems contribute to this process. The indices l and t refer to longitudinal and transverse phonons, respectively. For the limiting cases $\omega\tau \gg 1$ and $\omega\tau \ll 1$ analytical expressions can be derived but in general the integration has to be carried out numerically. The results of our numerical fits are shown in Figs. 2 and 3 by solid lines. Up to 5 K the resonant interaction [Eq. (1)] is dominant and perfect agreement with the experimental data is obtained for both velocity and attenuation. $n_0M_l^2=2.2\times10^7$ J/m^3 is deduced in good agreement with earlier measurements.[5,8] At higher temperatures the relaxation process [Eq. (3)] takes over and the quality of the approximation decreases. As usual we approximate the density of states by $n(E)=n_0[1+a(E/k)^2]$ (Ref. 9) where a is a free parameter. For our fit the following parameters were used: $a=0.0035$, $n_0D_l^2=2.6\times10^8$ J/m^3, and $\overline{M}=0.26$ eV, where $\overline{M}$ is the mean value of the coupling constants of Eq. (4)

$$\overline{M}^2=(M_l^2/v_l^5+2M_t^2/v_t^5)/(1/v_l^5+2/v_t^5) \quad .$$

These three parameters do not enter separately and therefore their choice is not without ambiguity. The most critical step lies in the choice of $\overline{M}$. From ultrasonic experiments—as for example from saturation and echo measurements[2,10] - $\bar{M} \simeq 1-2$ eV can be deduced for 2LS of small energy splitting ($E/k \simeq 0.04$ K). The choice of such a high value for $\bar{M}$ would cause an absorption maximum around 15 K and a shift of the maximum in the sound velocity to much lower temperatures in our fit. Our small value for $\bar{M}$ reflects the fact that the coupling parameter M is not a real constant, but varies even for 2LS having the same energy splitting.[2] In the resonant absorption the most efficient 2LS are those with the largest coupling constant. For the relaxation process, however, also 2LS with smaller coupling constants are important, reducing the mean value of $\bar{M}$. In the case of a, our best fit is obtained with a very weak curvature of the density of states. So far, higher values ($a = 0.01, \ldots, 0.078$) are reported,[5,8,9] but these values have been used to explain experiments at lower temperatures. Obviously, $n(E)$ does not increase as fast as expected going to higher level splitting of the 2LS. Finally $n_0 D_l^2$ deduced from our data is close to that known from previous experiments.[11]

The discrepancy between the theoretical fit and experiment at higher temperatures seems to be conceptional: If one of the two quantities—absorption or velocity of sound—is described correctly, the other quantity is overestimated or underestimated. Taking into account additional interactions, for example higher-order processes which are likely to become important at higher temperatures, does not improve the fit. On the other hand it is tempting to try to improve the agreement by introducing the tunneling model.[12] In this case the density of states $n(E)$ is replaced by two distribution functions; in addition the coupling parameters are energy dependent. But again the discrepancy stays, although it is always possible to fit either absorption or velocity perfectly well, even if the velocity variation is linear in temperature.[13]

V. CONCLUSION

For the first time we have carried out Brillouin scattering experiments in a solid at temperatures below 1 K. Even at these temperatures the propagation and attenuation of 35-GHz phonons in vitreous silica could be observed. The resonant absorption of these hypersonic phonons by 2LS has been determined, extending by more than one order of magnitude the frequency range of previous measurements.[14,15] In our experiments, the resonant attenuation is observed in the regime $\hbar\omega > kT$, and under strictly unsaturated conditions, due to the low intensity of phonons probed in light-scattering studies. We find that the measured absorption tends to a constant value at the lowest temperatures, as predicted by the theory.

Our measurement of the mean free path of 35-GHz phonons, which are dominant at 0.4 K, is in good agreement with that deduced from thermal conductivity at this temperature,[16] assuming that the mean free path is similar for longitudinal and transverse phonons in this temperature range.[17] Whereas only qualitative agreement between theory and experiment is found above 10 K, the low-temperature data are fully described by current theories based on the existence of two-level systems in glasses.

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