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Linear dispersion of transverse high-frequency phonons in vitreous silica

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Using phonon interference in thin films, we have measured the dispersion curve of transverse phonons in vitreous silica. A linear dispersion was found up to 450 GHz. This result has important implications for theories on the specific heat and thermal conductivity of glasses and is consistent with recent model calculations of phonons in a glass.

It is well known that glasses have universal low-temperature properties that differ from those of crystals.[1] The specific heat follows a $c_1T+c_3T^3$ law. The T term was successfully explained by a distribution of two-level systems.[2] The T^3 term is, at least in vitreous silica,[1,3] "excessive" in the sense that it is considerably larger than expected from the Debye model if ultrasonic velocities are used. The thermal conductivity, on the other hand, increases with T^2 below 1 K. This behavior was ascribed to phonon scattering from two-level systems.[2] Above 1 K a plateau is found in the thermal conductivity of all glasses studied so far, which is not yet understood satisfactorily.[4]

So far it has been impossible to assess the extent of the anomalies of the excessive T^3 term and of the plateau because both of them could be at least partly due to an unexpected softening of the transverse phonons. This was suggested recently by Jones, Jäckle, and Phillips.[5] The dispersion curve they needed to fit the excessive T^3 term in the specific heat produced also a plateau in the thermal conductivity.[5] As a further clue to the plane-wave-phonon nature of the states causing the excessive T^3 term appears the fact that they were found to be "fast" in time-dependent specific-heat experiments of Loponen *et al.*[6] Very recently, however, Grest, Nagel, and Rahman[7] published model calculations of the dispersion curves in a glass. They found no dispersion of the transverse phonons up to the largest-k vectors. In this situation, the experimental determination of the dispersion curve of real glasses is highly desirable. The traditional tool, neutron scattering, however, cannot be used near the center of k space in an amorphous solid.

In this Communication, we present the first direct measurements of the dispersion of transverse phonons in the relevant frequency range of several times 100 GHz. The data were obtained by observing phonon interferences[8] at thin films of vitreous silica. The phonons were generated and detected utilizing superconducting tunnel junctions.[9]

The experimental setup is shown as the inset in Fig. 1. A disk-shaped Si crystal, 4 mm thick, [100] oriented, was used as the base material. On one side of this crystal a 530-Å-thick oxide film was grown by dry oxidation at 1050 °C for 30 min. The thickness was measured by ellipsometry. The process conditions were those of metal-oxide-semiconductor fabrication. The resulting SiO_2 is structurally equivalent to vitreous silica quenched from the melt. It is homogeneous over the whole thickness[10] and the thickness of the Si-SiO_2 interface is of the order of only one atomic layer.[11] Atop of the SiO_2 film a tunnel junction made of a $Pb_{0.85}Bi_{0.15}$ alloy was placed. This junction is capable of generating monochromatic phonons with frequencies tunable up to about 700 GHz.[12] On the opposite surface of the crystal, an Al tunnel junction was prepared which served as detector for the phonon pulses of about 100-ns duration. Only transverse phonons were detected in the available samples due to the phonon-focusing conditions. Phonon-mode conversion at the Si-SiO_2 interface was

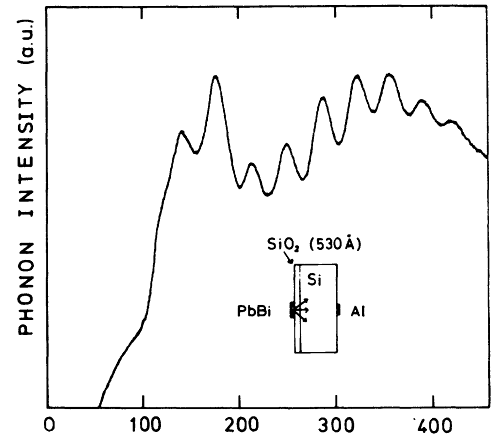

FIG. 1. Intensity of the phonon signal transmitted through a glass film acting as a Fabry-Perot interferometer. Inset: Sample setup.

negligible because of the near-normal incidence of the phonons. The glass film acts as a Fabry-Perot filter for the phonons. Whenever the phonon frequency hits one of its eigenstates with normal k, the transmission to the detector is maximum. These eigenstates correspond to $k_n = (\pi/d)n + k_0$, where d is the thickness, n an integer, and k_0 depends on the boundary conditions.

In Fig. 1, the measured phonon intensity is shown as function of frequency. At about 100 GHz there is a steplike increase of the signal strength because the phonon frequencies exceed the detector threshold $2\Delta_D/h$, the superconducting energy gap of Al. Above this frequency, the overall shape of the trace follows those observed in other crystals and depends on the monochromatic yield of the generator and the spectral response of the detector. Between 100 and 450 GHz, the phonon intensity is modulated by nine well-defined maxima and minima which must be due to phonon interference in the glass film. From the observability of an extremum at 450 GHz follows that the SiO_2 film was parallel within at least a quarter wavelength. With the sound velocity evaluated below, this amounts to 20 Å. Besides roughness, phonon scattering in the glass could also prevent the observation of higher-order extrema. From thermal-conductivity data Jäckle obtained at 450 GHz a mean free path[13] of about 400 Å which is close to our film thickness. Thus either effect could be dominant. We estimate, however, that the angular spread of the phonons has no detrimental effect up to frequencies of at least 600 GHz. It is interesting to note that from the observation of an interference at 450 GHz follows that the spectral width of the eigenstates in the film must be less than 5% at this frequency.

The frequencies of the maxima and the minima in-between are plotted as a function of k in Fig. 2. The zero point of the k scale is not known for the time being. It is obvious from Fig. 2 that all extrema lie perfectly on a straight line, which means that the dispersion is linear indeed. From the slope of the dispersion curve, we obtain the sound velocity of transverse phonons $v_t = (3700 \pm 120)$ m/s. This value is almost identical to the 3750 m/s obtained by ultrasonic techniques in the MHz frequency range.[14] It is therefore safe to assume that the dispersion is also linear between 1 and 100 GHz, where data are not available. This assumption sets k_0 to $(-0.50 \pm 0.05)\pi/d$. Figure 2 was already plotted this way. The observed orders n ranged from 4 to 12. Also shown in Fig. 2 is the dispersion curve necessary to fit the specific-heat data.[5,15] One can see that it is impossible to reconcile the nonlinear dispersion model with our data.

In another experiment we studied SiO_2 films prepared by electron-beam evaporation under UHV and high-vacuum conditions, respectively. Again, no dispersion was found between 100 and 400 GHz., The phonon velocities, however, were lower, namely, 3330 m/s in the UHV film and 2530 m/s in the high-vacuum film. These values were independent of film thickness which was varied between 350 and 600 Å. Evaporated films have a higher OH-ion content,[16] and we believe that it is possible that these impurities lead to an "opening" of the SiO_2 structure by breaking the Si—O—Si bonds and thereby reduce the stiffness of the solid. Actually, a reduction of the low-frequency sound velocity with increasing OH content was already found by Krause with ultrasonic techniques.[14] If we extrapolate his dependence of the sound velocity on the OH content over several orders of magnitude, we find that a few percent of OH in the SiO_2 are sufficient to reach our high-vacuum value which does not appear unreasonable.

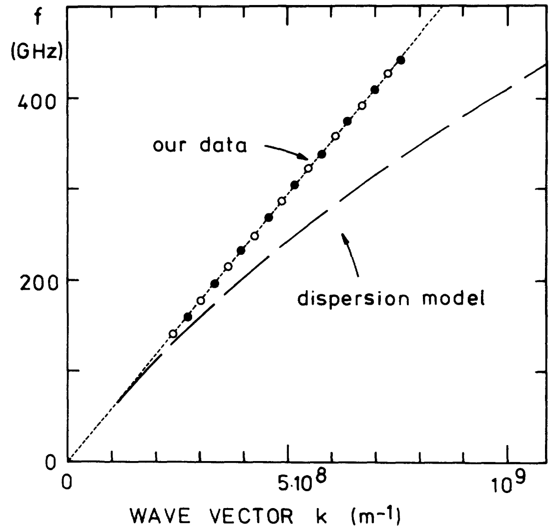

FIG. 2. Frequencies of the maxima (○) and minima (●) in Fig. 1 plotted vs the corresponding wave vector k. The short-dashed line is a linear fit to our data; the long-dashed line is the expected behavior from the nonlinear dispersion model (Ref. 5).

To conclude, we have measured the dispersion curve of transverse phonons in a frequency range which was hitherto not accessible by other techniques. The observed linear dispersion in vitreous silica rules out plane-wave phonons as a reason of the excessive T^3 term in the specific heat. This term must be caused by another kind of excitation. The coupling of these extra excitations to the phonons has to be rather strong to explain the absence of a time dependence of their specific heat.[6] The plateau in the thermal conductivity cannot be explained by nonlinear dispersion either. Rather, it must be caused by phonon scattering. Possibly the same extra excitations are also responsible for the plateau and for the strong inelastic phonon scattering observed recently.[17]

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