that the presence of an electron group with a lower temperature has an important effect on IAW and waves in which electron pressure is the dominant restoring force.

A possible interesting application might be to use a cold-electron source to stabilize IAW instabilities via Landau damping due to the expected reduction of the wave phase velocity by the cold electrons. By the same token, such manipulation of the IAW dispersion by the introduction of a cold-electron component could lead to rapid ion heating due to preferential wave energy deposition into the ions.

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†Permanent address: Ecole Polytechnique, Paris, France.

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Temperature Dependence of the Far-Infrared Absorption Spectrum in Amorphous Dielectrics*

K. K. Mon, Y. J. Chabal, and A. J. Sievers
Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University,
Ithaca, New York 14853

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A study of the temperature dependence of the absorption coefficient in amorphous dielectrics below 10 cm^{-1} has enabled us to identify two level configurations from thermal population changes. We also find that the anomalous low-frequency modes in glasses consist of widely spaced manifolds of energy levels.

Measurement of the thermal properties of glasses at low temperatures has provided experimental evidence that anomalous low-frequency modes occur in amorphous materials.[1,2] The linear temperature dependence of the specific heat which occurs in glasses below a few degrees kelvin is interpreted as arising from these anomalous modes whose frequencies lie below 4 cm^{-1}. Both the nonlinear ultrasonic attenuation[3] around 0.1 cm^{-1} and the anomalous sound-velocity measurements[4] around 0.01 cm^{-1} at very low temperatures have established that the modes at those frequencies are very anharmonic and that the density of modes is consistent with the specific-heat measurements.[1,2] In addition to these special modes, Brillouin-scattering experiments[5] have shown that there are regular well-behaved transverse and longitudinal phonons in this frequency region as well. To account for these special modes a tunneling model has been proposed[6,7] in which atoms, ions, or groups of particles quantum mechanically tunnel between two or more equivalent sites. The linear dependence of the specific heat has been obtained from this model by including a statistical distribution of barrier heights and asymmetries for the local potential. One straightforward prediction which this model makes is that the temperature dependence of the far-infrared absorption coefficient should be determined by the occupation numbers of a small number of energy levels associated with the tunneling manifold. For example, a distribution of infrared-active two-level systems will produce an infrared absorption which decreases with increasing temperature in a well-defined way.

In this Letter, we describe the first measurements of the temperature-dependent absorption coefficient of amorphous dielectrics in the far infrared at frequencies between 2 and 10 cm^{-1}. We find the decrease in absorption expected for two-level systems in a restricted temperature and frequency range. Our more detailed measurements show a complex behavior which we believe points to the existence of excited-state transitions in the far-infrared spectral region.

For the spectroscopic measurements, we have used a lamellar-grating interferometer together with a liquid-He^3–cooled germanium bolometer.[8] Four different glasses have been studied, namely SiO_2, SiO_2 with 600 ppm H_2O, GeO_2 between 1.2 and 4.2°K, and polymethylmethacrylate (PMMA) (Plexiglas) between 0.54 and 10°K. The PMMA samples were obtained from commercial stock. Both SiO_2 (Optosil) and SiO_2 with 600 ppm H_2O (Infrasil II) were obtained from Valprey Corporation while the GeO_2 samples were prepared at the Cornell University crystal-growing facility. The absorption coefficients were obtained from comparing transmission measurements on samples of many different lengths. For the SiO_2 measurements lengths up to 54 cm were used. The incident power on the samples was estimated to be less than 10^{-9} W.

The absorption coefficient of GeO_2, PMMA, vitreous SiO_2 with 600 ppm H_2O, and vitreous SiO_2 at 4.2°K is shown in Fig. 1. Our experimental measurements below 10 cm^{-1} in Fig. 1 demonstrate that some structure in the frequency dependence of the absorption coefficients exists at low temperature. This is very different from results reported by Strom *et al.*[9] at room temperature.[10]

The temperature range of interest is less than 10°K. Experimentally we have measured $\Delta\alpha(\omega, T) = \alpha(\omega, T) - \alpha(\omega, T_0)$, where the lower temperature is defined as T_0. The absorption coefficient was measured at many intermediate temperatures but for clarity Fig. 2 is restricted to experimental measurements at the temperature extremes or to regions which illustrate different characteristic features.

Figures 2(a) and 2(d)–2(f) show the change in absorption coefficient between 1.2 and 4.2°K for the four different glasses. The similarity of the frequency dependence in each case is quite striking. With increasing temperature near 4.2°K for frequencies less than 6 cm^{-1} ($\hbar\omega/2kT < 1$) the ab-

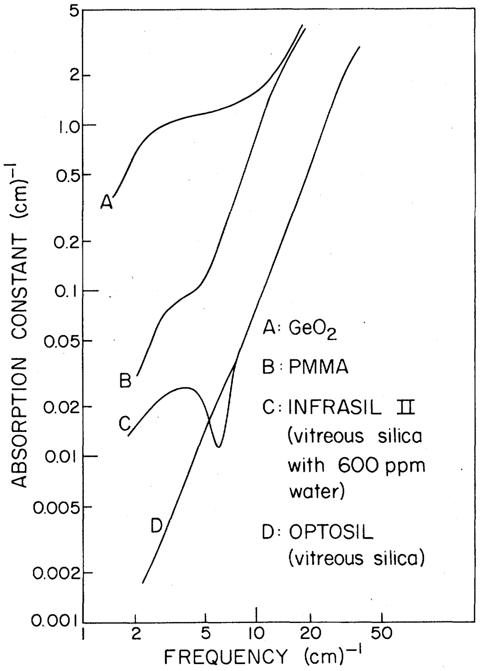

FIG. 1. Absorption coefficient versus frequency for four different glasses at 4.2°K.

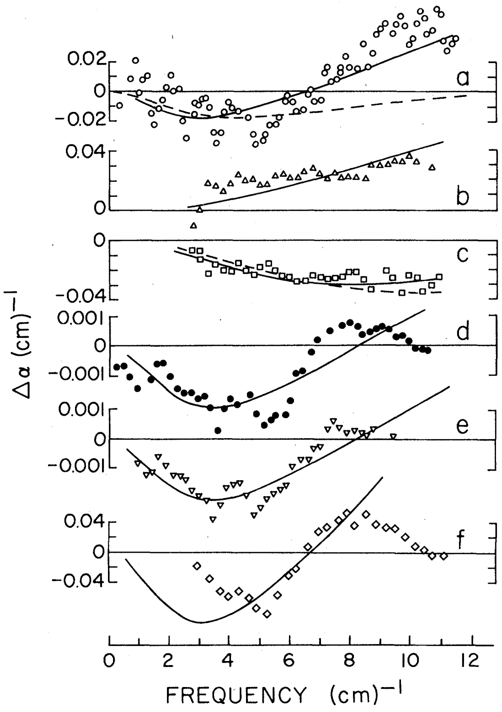

FIG. 2. Changes in absorption coefficient with temperature versus frequency. The dots represent the experimental data, the dashed line the two-level model, and the solid line the three-level model. (a) PMMA: $\alpha(4.2°K) - \alpha(1.2°K)$. Parameters for the three-level model are $g_0 = 3.6\times10^{-2}$ and $r = \frac{1}{20}$. (b) PMMA: $\alpha(0.97°K) - \alpha(0.55°K)$. (c) PMMA: $\alpha(8.68°K) - \alpha(4.25°K)$. (d) SiO_2 with 600 ppm H_2O: $\alpha(4.2°K) - \alpha(1.2°K)$. Parameters for the three-level model are $g_0 = 3.8\times10^{-3}$ and $r = \frac{1}{40}$. (e) SiO_2: $\alpha(4.2°K) - \alpha(1.2°K)$. Parameters for the three-level model are $g_0 = 3.8\times10^{-3}$ and $r = \frac{1}{40}$. (f) GeO_2: $\alpha(4.2°K) - \alpha(1.2°K)$. Parameters for the three-level model are $g_0 = 2\times10^{-1}$ and $r = \frac{1}{20}$.

sorption coefficient decreases with increasing temperature while for frequencies greater than 6 cm^{-1} ($\hbar\omega/2kT > 1$) the absorption coefficient increases with increasing temperature.

Figure 2(b) shows that near the low-temperature extreme (0.54°K) we observe an increase in absorption coefficient with increasing temperature throughout our frequency range ($\hbar\omega/2kT > 1$).

Finally near 8°K at the high-temperature extreme over our frequency range ($\hbar\omega/2kT < 1$) we show in Fig. 2(c) that an increase in temperature produces a decrease in the absorption coefficient.

The decrease in absorption coefficient with increase in temperature can be described quantitatively for all the glasses by the thermal population dependence of a simple two-level model. The absorption coefficient associated with a statistical distribution of optically active two-level systems (level spacing $\hbar\omega$) and a temperature-independent absorption $H(\omega)$ produced by defect-induced processes in regular harmonic phonons can be written quite generally as

$$\alpha(\omega, T) = G(\omega)\tanh(\hbar\omega/2kT) + H(\omega). \qquad (1)$$

We determined $G(\omega)$ at each frequency by plotting the experimental $\Delta\alpha(\omega, T)$ versus $\tanh(\hbar\omega/2kT) - \tanh(\hbar\omega/2kT_0)$ for temperatures near 4.2°K. Good fits by straight lines have been obtained in the low-frequency region below 6 cm^{-1}. The slope of such a line gives $G(\omega)$ at one frequency. Because ω, T, and T_0 are all measured quantities there are no experimental parameters in this fit. The dipole moment estimated from our $G(\omega)$ is consistent with the dipole moment estimated by von Schickfus, Hunklinger, and Piché.[11]

Such a model cannot explain the temperature dependence of the absorption coefficient over the entire frequency range measured. For example the dotted line in Fig. 2(a) shows the predicted absorption coefficient for a two-level system. With increasing temperature the absorption coefficient always decreases and the $\Delta\alpha$ for this model must remain negative at all frequencies.

The PMMA data in Figs. 2(a)–2(c) show the inadequacy of the two-level model: The model works below 6 cm^{-1} in 2(a), does not even give the correct sign for the low-temperature extreme in 2(b), and fits the high-temperature extreme in 2(c) over the entire frequency region studied.

One possible explanation of the deviation from the two-level description would be to introduce an additional absorption mechanism associated with relaxation processes proportional to T.[12] We have attempted to fit the data in Figs. 2(a) and 2(b), and additional low-temperature data,[13] with a T^n term added to the absorption coefficient in Eq. (1). We find that the data restrict n to an unphysically small value ($\lesssim 0.1$) and so we discard this possibility.

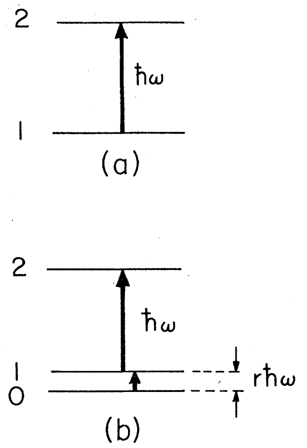

FIG. 3. Two models used in the text: (a) two-level; (b) three-level. The far-infrared transition is labeled $\hbar\omega$.

To account for the temperature dependence of the absorption coefficient over the entire frequency range we have incorporated the two-level model described above into a three-level scheme. The energy-level diagram is shown in Fig. 3. Two transitions are allowed: $\hbar\omega$ and $r\hbar\omega$. The density of states is assumed to be constant up to 20 cm^{-1} and then to drop abruptly to zero. This cutoff frequency is well above the anomalous frequency region determined from specific heat. For $r \ll 1$ only the excited-state transition ω is in our frequency range.

The absorption coefficient associated with this excited-state transition is

$$\alpha(\omega, T) = \omega g_0 \frac{1 - \exp(-\hbar\omega/kT)}{1 + \exp(r\hbar\omega/kT) + \exp(-\hbar\omega/kT)} + H(\omega), \qquad (2)$$

where $g_0 = (8\pi^3/3ch)|P|^2 n_0$. If we express ω in inverse centimeters then g_0 is dimensionless. P is the electric dipole matrix element and n_0 is the density of modes per inverse centimeter. This model reduces to an effective two-level model near 4.2°K below 6 cm^{-1} with $r \ll 1$. g_0 is determined from the 4.2°K data while r is obtained from lower-temperature measurements. The values of g_0 and r are given for each of the glasses in Fig. 2. The solid curves in Fig. 2 illustrate the three-level–model fits to the experimental data. The fit for GeO_2 in Fig. 2(f) is satisfactory only near 6 cm^{-1}, but the general behavior is still accounted for.

The three-level scheme appears to contain the essential physical features observed over the entire temperature and frequency range. Our fit is not unique and no doubt better agreement with the data could be obtained with a four-level model and an additional frequency-dependent correction to the assumption of constant density of states, especially in GeO_2 and SiO_2 near 10 cm^{-1}. We do not claim that the low-temperature modes in glasses consist of three levels; no doubt the individual manifolds are quite intricate, but the success of the three-level model described here shows for the first time that the anomalous low-frequency modes in these glasses do contain widely spaced manifolds of energy levels.

One last check on the consistency of this model can be made by comparing an estimate of the dipole matrix element obtained here for SiO_2 with that obtained by temperature-dependent dielectric-function measurements of vitreous silica.[11] Using our values of $g_0 = 3.8\times10^{-3}$ and $n_0 \sim 2\times10^{17}$/cm^{-1} cm^3 from specific-heat measurements,[2] we obtain for the dipole matrix element $P\sim(0.04\ \text{Å})e$. This is comparable to the value $P\sim(0.063\ \text{Å})e$ obtained for the vitreous silica.[11]

In conclusion, we have shown that thermal population effects can be observed in glasses in the far infrared. In addition we have found that the complex temperature dependence and frequency dependence observed in four different glasses can be understood in terms of simple two- and three-level models. Such a distribution of levels has already been observed spectroscopically[14] for ion tunneling in single crystals and points to a tunneling configuration for the low-frequency anomalous modes in glasses.

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