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Nonlinear Phonon Propagation in Fused Silica below 1 K

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Phonon pulse propagation has been studied in fused silica at frequencies between 0.4 and 2.0 GHz for temperatures as low as 0.1 K. An increase in attenuation, observed as the incident acoustic intensity is decreased, is attributed to resonant scattering by two-level "tunneling states" intrinsic to glasses.

Recent experimental observations on a variety of insulating glasses have revealed several anomalous properties at temperatures T below 1 K. In particular, the specific heats are large (linear in T) relative to pure crystalline insulators, while thermal conductivities are found to be unusually small.[1] Using a kinetic formulation of heat transport, Zeller and Pohl[1] extracted from their thermal-conductivity data an average phonon mean free path roughly proportional to T^{-1} and anomalously small; e.g., $\bar{l} \approx 1.5 \times 10^{-2}$ cm for fused SiO_2 at 0.1 K. This result for $\bar{l}$ is, however, in apparent disagreement with more direct measurements of phonon decay lengths in fused silica by stimulated Brillouin emission[2,3] and ultrasonic studies[3] which have yielded decay lengths 1–3 orders of magnitude larger than Zeller and Pohl. It has been suggested that the anomalous specific heat may arise from a distribution of localized two-level "tunneling defects" intrinsic to the glassy state and that the short thermal mean free paths are the result of resonant scattering of phonons by these two-level systems.[1,4-6] The discrepancy in the magnitudes of the mean free paths can then be understood, if the direct experiments were performed at phonon intensities sufficiently great for saturation of the two-level systems to occur.[2,5]

In the present paper, we present experimental evidence which offers support for the tunneling-state model of the low-temperature thermal properties of glasses. We have measured the attenuation of phonon pulses in fused silica at temperatures from 0.1 to 2.5 K, for frequencies between 0.4 and 2.0 GHz, and for pulse energies ranging from 10^{-1} to 10^{-7} erg/cm^2. At the high end of our frequency range, for $T \lesssim 0.5$ K, we indeed observe a marked decrease in the phonon path length as the incident phonon pulse energy is reduced below about 10^{-3} erg/cm^2. Our observations of the temperature, frequency, and energy dependence of this effect are in general agreement with a calculation of the response of two-level tunneling states to "intense" phonon pulses.

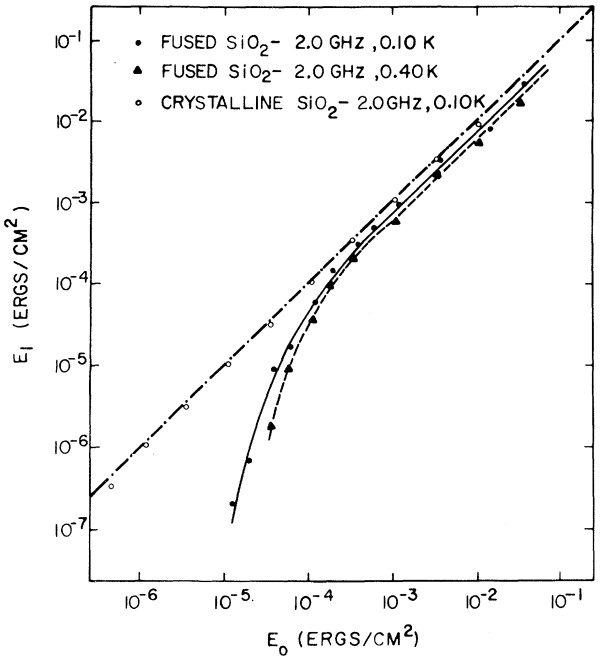


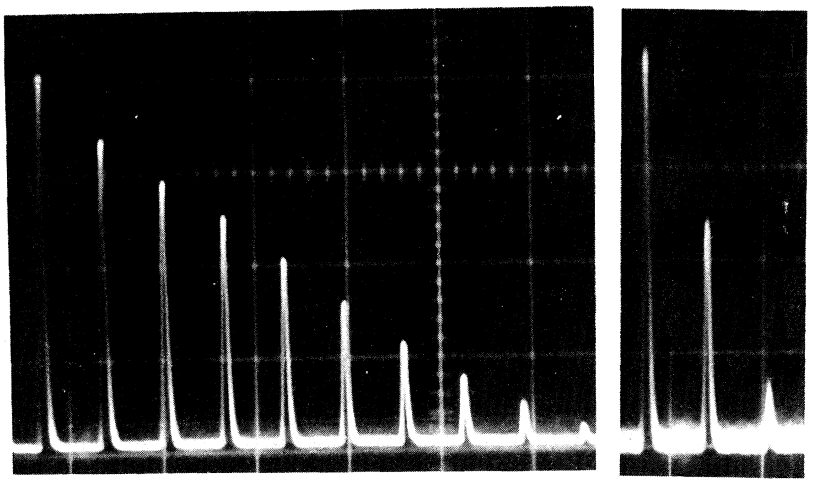
FIG. 1. Acoustic energy per square centimeter in the first echo, E_1, as a function of initial acoustic energy per square centimeter, E_0, per 0.5-μsec pulse. Propagation distance in fused SiO_2 is 3.80 cm; in Z-cut quartz, 1.46 cm. Dot-dashed line represents lossless propagation.

Experimental results obtained at 0.10 K are plotted in Fig. 1 for 2.0-GHz longitudinal phonon pulses of duration $\tau = 0.5$ μsec. The measured energy per square centimeter in the first phonon echo, E_1, corresponding to a propagation distance $2L$ of 3.8 cm, is plotted as a function of the energy per square centimeter of the initial pulse, E_0. At sufficiently large energies the medium is linear, with an effective energy decay length given by $l_{sat} = 2L[\ln(E_0/E_1)]^{-1} \approx 15$ cm. We attribute the measured l_{sat} to inhomogeneity scattering, to beam diffraction, and to temperature-dependent relaxation processes[4,5] (i.e., nonresonant absorption by the two-level systems). As the incident energy is reduced below $\sim 10^{-3}$ erg/cm^2 in SiO_2 *glass*, the echo energy drops very rapidly in a highly nonlinear way. In contrast, the behavior of 2.0-GHz phonons propagating in *crystalline* SiO_2 (Z-cut quartz) is commensurate with the response of a lossless linear medium, within our experimental accuracy. The magnitude of the nonlinear absorption in the glass depends upon the pulse width, decreasing as the width increases. The decay rate is independent of repetition frequency, however, and was unchanged when two successive pulses were injected, separated by as little as ~ 0.2 μsec. The excess absorption is nearly temperature independent, as shown by the results at 0.4 K in Fig. 1, although the background absorption (l_{sat}^{-1}) increases markedly with T above 0.4 K. There is, however, a strong frequency dependence since at 0.85 GHz large nonlinearity is apparent only at energies ≈ 50 times smaller than at 2.0 GHz, and at 0.43 GHz linear behavior is observed at all measurable energies.

Echo decay patterns at 1.9 GHz and 0.1 K are shown in Fig. 2 for two values of E_0, 1.5×10^{-2} erg/cm^2 in (a), and 5×10^{-4} erg/cm^2 in (b). The decrease in the decay time in (b) relative to (a) is quite apparent although the deviation from linearity of the first echo is only slightly visible on the logarithmic scale of Fig. 1. Note also that the echo amplitude does not decay exponentially with time but exhibits a nearly linear dependence on time, consistent with an energy decay rate $\propto E^{-1/2}$.

The experiments were performed in a He^3-He^4 dilution refrigerator contained within a superconducting solenoid.[7] The specimens were Suprasil fused silica, dimensions 1.91 cm$\times$0.64 cm$\times$0.64 cm, with the small faces polished to a laser finish. One polished face was coated, in sequence, with Cr and Au films for a ground plane, a sputtered ZnO film which served as transducer, and a top Cr and Al electrode which terminated a miniature superconducting coaxial transmission line.[8] The single transducer served as transmitter and receiver for the phonon pulses and their echoes. The structure was nonresonant and exhibited a relatively flat response from 0.4 to 2.3 GHz. Identical results were found in runs near 2 GHz for two Suprasil samples having ZnO films which differed in thickness by about a factor of 2. The acoustic transfer function was determined by a pulse comparison method, in which the amplitude of an echo was compared (to within ± 0.5 dB) to a delayed reference pulse inserted into the transmission line before the microwave receiver. This method eliminates receiver nonlinearities and allows a calibration of the peak power in the received echoes. A Ge thermometer, mounted on the sample, was used to determine the absolute temperature and a Speer $\frac{1}{2}$-W 470-Ω carbon resistor, mounted on the same face, provided a secondary thermometer of high sensitivity near 0.1 K.[9]

According to the tunneling-state model for the anomalous properties of glasses below 1 K, the



(a) (b)

FIG. 2. Photographs of detected echo amplitudes in fused SiO_2 at 0.1 K for 1.9-GHz, 0.5-μsec longitudinal phonon pulses; (a) $E_0 = 1.5\times10^{-2}$ erg/cm^2; (b) $E_0 = 5\times10^{-4}$ erg/cm^2. Note the increased decay rate for lower incident energy in (b) and the nonexponential time dependence. The amplitudes of first echos are normalized to the same height. Time scale (abscissa): 5 μsec/(major division).

levels most important for phonon absorption are those for which the two wells are nearly symmetric at equilibrium.[4-6] For simplicity, we shall consider the exactly symmetric case ($\lambda = \lambda_{min}$, in the notation of Ref. 4). The response of the two-level system to the incident phonon field is similar to the behavior of spins in the electromagnetic field in a spin-resonance experiment or a maser.[10,11] Let us write the density matrix ρ_s for the two-level system in the form $\rho_s = (1 + \vec{p}\cdot\vec{\sigma})/2$, where $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are the Pauli "spin" matrices. The equation of motion for $\vec{p}$ has the form[10,11]

$$d\vec{p}/dt = \vec{p}\times\vec{h} - \hat{x}\Gamma p_x - \hat{y}\Gamma p_y - 2\hat{z}\Gamma(p_z - p_0), \quad (1)$$

where $p_0 \equiv -\tanh(\epsilon/2k_BT)$ is the equilibrium "polarization," $\vec{h} \equiv (2\eta\hbar^{-1}, 0, -\epsilon\hbar^{-1})$ is the "effective magnetic field," ϵ is the equilibrium splitting of the two levels, and the perturbation $\eta = \mathrm{Re}(\eta_0 e^{-i\omega t})$ is proportional to the local strain field of the incident phonon pulse. The amplitude η_0 is related to $\mathcal{E}_i$, the energy density in the incident pulse, by $|\eta_0|^2 = 2\mathcal{E}_i\gamma_i^2/\rho c_i^2$, where ρ is the mass density, and c_i and γ_i are the sound velocity and the deformation potential constant for the incident mode, respectively. The relaxation rate Γ arises primarily from emission and absorption of transverse phonons, so that

$$2\Gamma|p_0| = \Gamma_0 \approx \gamma_t^2\epsilon^3/\pi\rho\hbar^4c_t^5, \quad (2)$$

where Γ_0 is the transition rate from the upper to the lower state at 0 K, c_t is the transverse sound velocity, and γ_t is an "average" deformation potential coefficient for transverse modes.[4,5]

If the phonon field strength η_0 is known as a function of time, Eq. (1) may be solved for $\vec{p}(t)$, and one may then compute the energy absorbed by the two-level system. For resonant absorption of the incident phonon, we are interested in the case in which $|\hbar\omega - \epsilon_0|$ and η_0 are small compared to ϵ. A decay rate is computed by adding up the losses due to all tunneling levels in a unit volume, and dividing by the incident energy flux. We assume that $\tilde{n}(\epsilon)$, the number of contributing levels per unit volume and unit energy ϵ, is a slowly varying function of ϵ, and we shall make the further simplifying approximation that Γ_0 and γ_i are the same for all of these level pairs. Two limits may be easily evaluated. If $\eta_0\hbar^{-1}$ is small compared to *either* Γ or τ^{-1}, the energy absorbed is linear in the incident intensity and the phonon decay length is given by $l^{-1} = l_0^{-1}\tanh(\hbar\omega/2k_BT)$, where l_0 is the mean free path at $T = 0$ K:

$$l_0^{-1} = \pi\tilde{n}(\hbar\omega)\gamma_i^2\omega/\rho c_i^3. \quad (3)$$

On the other hand, for strong incident intensities such that $\eta_0\hbar^{-1}$ is larger than Γ and τ^{-1}, nonlinear (saturation) effects occur. In the cw limit ($\tau \gg \Gamma^{-1}$), we find after some computation the following result for the differential rate of decay of the acoustic energy with distance:

$$l^{-1} \equiv -c_i^{-1}d(\ln E)/dt = l_0^{-1}(\mathcal{E}_c/\mathcal{E})^{1/2}, \quad (4)$$

where

$$\mathcal{E}_c \equiv \hbar^2 \Gamma_0^2 \rho c_i^2 / 4\gamma_i^2, \quad (5)$$

and $\mathcal{E}$ is the "average" density in the pulse. (More accurately, $\mathcal{E}^{-1/2}$ is the integral over space of $\mathcal{E}_i^{1/2}$, divided by the total energy in the pulse.) This form for the absorption occurs because the levels with $|\epsilon - \hbar\omega| \lesssim \eta_0$ are saturated by the pulse, and each of these levels dissipates energy at the rate $\epsilon\Gamma_0$. Equation (4) disagrees with the calculation of Jäckle,[5] who finds $l^{-1} \propto \mathcal{E}^{-1}$, for the resonant absorption at high power levels.

For a shorter pulse ($\tau \ll \Gamma^{-1}$ but still $\gg \hbar\eta_0^{-1}$), the situation is more complicated. In this case, however, the nonlinear attenuation will be accompanied by a marked pulse broadening,[10] which we do not observe in our experiments. The observation that successive pulses do not interfere also suggests $\tau \gtrsim \Gamma^{-1}$.

Equation (4) agrees well with most features of the nonlinear absorption observed experimentally. The effective decay length is correctly predicted to be proportional to $\mathcal{E}^{1/2}$, to be independent of T, and to be a strong function of frequency. [Specifically, Eq. (4) yields $l^{-1} \propto \omega^4$, provided γ_i and $\tilde{n}(\hbar\omega)$ are independent of ω.] However, our experimental result that l^{-1} varies with pulse duration disagrees with (4), and is difficult to understand if $\tau \gtrsim \Gamma^{-1}$.

The observed order of magnitude for l^{-1} in the nonlinear regime is consistent with reasonable estimates of the parameters in the model. Let us assume that $\gamma_i \approx 3$ eV, and that l_0 is the same for longitudinal and transverse phonons, with $l_0 \approx 0.1$ cm at 2 GHz.[1] Then Eqs. (2), (3), and (5) yield $\Gamma_0 \approx 10^6$ sec^{-1}, $\tilde{n}(\epsilon) \approx 2\times10^{18}$ states/eV cm^3,[12] and $\mathcal{E}_c \approx 2\times10^{-9}$ erg/cm^3. Equation (4) then predicts $E_0^{1/2} - E_1^{1/2} \approx 0.5\times10^{-3}$ (erg/cm^2)$^{1/2}$ for the pulses in Fig. 1, whereas the experimental results are described by $(3.0 \pm 1.5)\times10^{-3}$ (erg/cm^2)$^{1/2}$.

We see that the large nonlinearity in phonon pulse propagation observed in fused SiO_2 can be understood in terms of interactions with a distribution of resonant scatterers intrinsic to the glassy state—a model proposed earlier to explain the anomalous specific heat and thermal conductivity of glasses at very low temperatures. Continued study of the decay length at different power levels, temperatures, pulse durations, and frequencies, should enable one to determine the relaxation rate more precisely and to learn something about the statistical distribution of these levels.

The authors are grateful for helpful discussions with M. Lax and S. L. McCall.

Added note.—We have been informed by Dr. S. Hunklinger that an amplitude-dependent attenuation of longitudinal sound waves has also been observed in vitreous silica at low temperatures by S. Hunklinger, W. Arnold, St. Stein, R. Nava, and K. Dransfeld (to be published).

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[8]We are deeply grateful to N. F. Foster for preparing the highly efficient transducers. Also, we thank S. Bortas for polishing the crystal quartz sample.

[9]We thank K. Andres for kindly providing us with the calibrated Ge thermometer.

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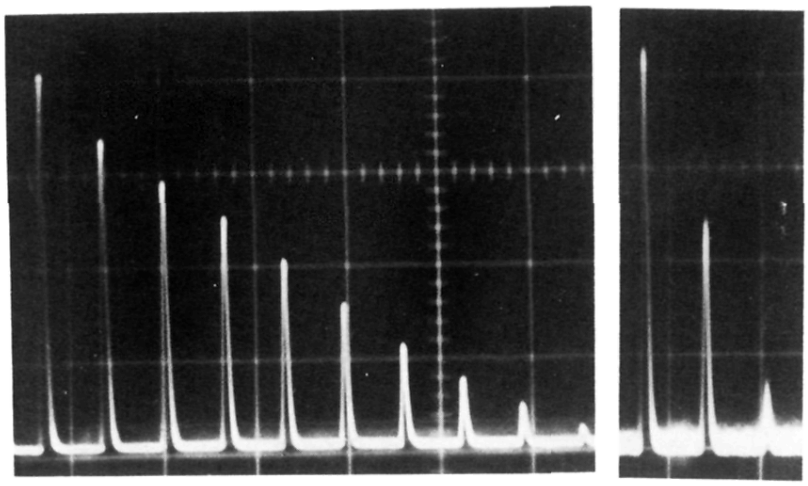

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