

exists below which the turbulence does not persist.

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Thermal Relaxation of Low-Energy Excitations in Vitreous Silica

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Strong thermal-relaxation effects have been observed in vitreous silica at low temperatures on a time scale of order 10^3 s, which are attributed to slowly relaxing low-energy excitations intrinsic to the amorphous state. With the use of a simplified version of the tunneling model, a density of states is derived for these excitations, which is comparable in magnitude to the density of the fast-relaxing excitations derived from phonon-scattering experiments.

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Glasses below 1 K show anomalous thermal properties¹ which point to the existence of low-energy excitations (LEE) specific to the amorphous state. The theoretical model most widely accepted for the explanation of these properties is the tunneling model of Anderson, Halperin, and Varma,² and Phillips.³ This model connects the LEE with two-level systems which originate from the tunneling of as yet unknown atoms or molecular complexes through an energy barrier between two almost equivalent sites. However, the model in its standard form has not been able to cover the fast-growing body of experimental evidence with one unique set of parameters and thus has been under discussion.⁴ The existence of LEE in the amorphous state raises the question on what time scales these LEE will equilibrate with the phonon bath. According to the standard tunneling model, the LEE-phonon relaxation times should be widely distributed and the contribution of the LEE to the specific heat should depend on the measurement time.² The experimental evidence

to this point is at present conflicting: Measurements of heat-pulse propagation in vitreous silica and polymethyl methacrylate (PMMA) by Goubau and Tait⁵ on a 100- μ s time scale and in vitreous silica by Kummer, Dynes, and Narayanamurti⁶ on a 1- μ s time scale did not show the expected results, whereas Loponen *et al.*⁷ recently observed relaxation effects in specific-heat measurements on vitreous silica using spatially uniform heat pulses in small samples. From the latter results, an upper limit of about 10 ms has been derived⁷ for the LEE-phonon relaxation times which is considerably lower than that usually assumed in the tunneling model. In view of these discrepancies we have designed an experiment which specifically probes thermal relaxation effects on large time scales, here 50–6000 s. We have applied it to Suprasil W,⁸ a well-characterized pure type of silica glass, which has also been used in the relaxation work of Kummer, Dynes, and Narayanamurti⁶ and Loponen *et al.*,⁷ and we shall put forth strong evidence that there

is significant thermal relaxation in Suprasil W on these long time scales.

In our experiment the sample was weakly connected to a bath at constant temperature $T_0 = 0.200$ K. It was heated up to some initial temperature T_i between 0.205 and 3.52 K, thermalized at T_i , and then (at $t=0$) allowed to relax back to T_0 . The temperature decay was recorded and could be converted to the time-dependent heat release of the sample by means of the known thermal resistance between sample and bath.⁹

The sample was a cylindrical rod of 1.5 cm diameter and 10.1 cm length. Its temperature was measured with a thin slice of carbon resistor (Speer 100 Ω , $\frac{1}{4}$ W) by using an ac bridge at 1035 c/sec with a resolution of ± 0.05 mK. The thermal link to the bath, here the mixing chamber of a dilution refrigerator, consisted of a 6-cm length of 0.008-cm-diam copper wire. Its heat conduction $K = 6.19$ erg s⁻¹ K⁻¹ was measured in a steady-state heat-flow experiment. The bath temperature was regulated at $T_0 = 0.200$ K with a relative stability of about 2×10^{-4} /h. Thermal link, thermometer, and heater (Pt-W wire) were thermally anchored to the sample by means of three silver foils of about 3 cm² each and Apiezon N grease as described by Lasjaunias *et al.*¹⁰ The sample was completely surrounded by a radiation shield at mixing-chamber temperature and carefully shielded against vibrational and electromagnetic heating. Additional experiments, including the use of a second thermometer on the sample, were done to make sure that the internal time

constant of the sample assembly remained < 1 s in all cases.

Figure 1 shows the temperature decay of the sample recorded between 0.205 and 0.200 K for four different temperatures T_i . The right-hand scale displays the corresponding heat release \dot{Q} from the sample. The resolution in terms of power is 0.3×10^{-3} erg s⁻¹. The curve, where the sample was heated up to only $T_i = 0.205$ K, represents the usual relaxation-type measurement of the specific heat on a 10-s time scale. Within the limits of our resolution the decay is an exponential with a time constant of 18.1 s independent of thermalization time. Our result $c(T_0, t=10 \text{ s}) = 4.0$ erg K⁻¹ cm⁻³ is in good agreement with the published data¹¹: The curves originating at higher temperatures are nonexponential. They are reproducible within the limits of our resolution, provided that the sample had been thermalized at T_i for a sufficiently long time (e.g. 10 h for $T_i = 3.52$ K).

The measured energy release \dot{Q} of Fig. 1 consists of two parts:

$$\dot{Q}(T_i, t) = \dot{Q}_0(T_i, t) + \dot{Q}_1(T_i, t). \quad (1)$$

$\dot{Q}_0(T_i, t) = c(T(t), t=10 \text{ s}) V dT(t)/dt$ is the trivial contribution from phonons and all LEE with relaxation times < 10 s, i.e., all excitations which contribute to the specific heat measured on a 10-s time scale. (V is the volume of the sample.)

$\dot{Q}_1(T_i, t)$ is the extra contribution which we attribute to LEE with relaxation times > 10 s. Since we are interested in the latter, we calculate the trivial contribution and subtract it from the measured energy release $\dot{Q}(T_i, t) = K [T(t) - T_0]$ in accordance with Eq. (1). The calculation is done using our value of $c(T_0, t=10 \text{ s})$ for the limited temperature range 0.205–0.200 K considered and with $dT(t)/dt$ derived from the decay curves by means of least-squares approximations by cubic splines. $\dot{Q}_1(T_i, t)$ should vanish for samples without long-time relaxation effects. This was verified with a copper sample by using the same apparatus and addenda. The \dot{Q}_1 -vs- t curves resulting from this procedure are plotted in Fig. 2 along logarithmic scales, and this representation shows that the long-time heat release of the sample roughly follows a t^{-1} dependence for all thermalization temperatures T_i .

To compare our data with the results of other workers we tentatively apply the standard tunneling model of Anderson, Halperin, and Varma² and Phillips³ to the curves of Fig. 2. The two-

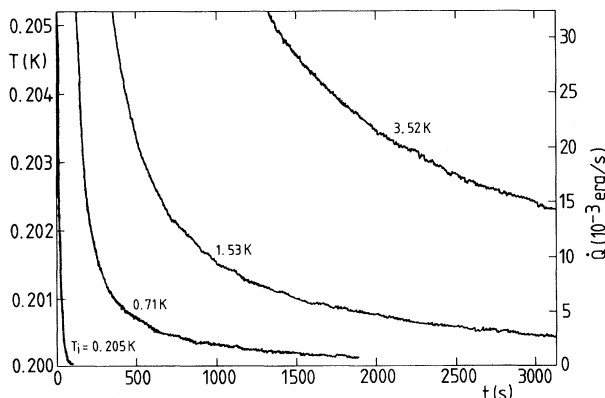


FIG. 1. Temperature decay (left scale) and power release (right scale) of a 17.9-cm³ sample of Suprasil W between $T = 0.205$ K and $T_0 = 0.200$ K as a function of time. Four typical curves with different T_i are shown. The sample was thermalized at T_i and at $t = 0$ allowed to relax back to T_0 .

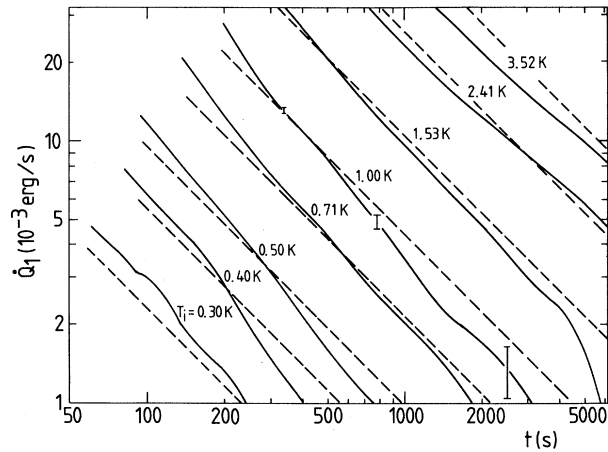


FIG. 2. Power \dot{Q}_1 released from slowly relaxing ($\tau > 10$ s) low-energy excitations in Suprasil W as a function of time. Eight curves with different T_i are shown. The typical measurement accuracy is indicated in one curve only. The broken lines represent t^{-1} dependences and are explained in the text.

level systems of this model have an energy splitting $E = (\Delta^2 + \Delta_0^2)^{1/2}$, where Δ is the asymmetry energy of the two potential wells in the absence of tunneling and $\Delta_0 = \hbar\omega_0 e^{-\lambda}$ is the coupling energy. $\hbar\omega_0$ is a typical zero-point energy in either well and is assumed constant. λ describes the extent of wave-function overlap between the lowest states in either well. The number of two-level systems per volume is assumed to be uniformly distributed as a function of Δ and λ : $P(\Delta, \lambda) d\Delta d\lambda = \bar{P} d\Delta d\lambda$. Coupling to the phonons leads to relaxation times, which for a given energy splitting E , vary over many orders of magnitude from a lower limit τ^* for the most strongly coupled systems to some upper limit τ_{\max} for a maximum barrier height of ~ 1 eV connected with the activation energy for diffusion. Within this standard tunneling model, the time-dependent specific heat for measuring times $\tau^* < t < \tau_{\max}$ is given by⁴

$$c(T, t) = \frac{1}{12} \pi^2 k_B^2 T \bar{P} \ln(4t/\tau^*). \quad (2)$$

Equation (2) may be used as starting point for the discussion of our curves, if we make the assumption that the phonon bath immediately cools down from T_i to T_0 at $t=0$ and that the energy, therefore, is released at constant bath temperature T_0 . For a sample of volume V we then find from Eq. (2), after differentiation with respect to t and integration with respect to T ,

$$\dot{Q}_1(T_i, t) = \frac{1}{24} \pi^2 k_B^2 \bar{P} V (T_i^2 - T_0^2) t^{-1}. \quad (3)$$

In the actual experiment the temperature does

change during the decay. However, concerning the temperature deviation during the tails of the decay curves (Fig. 1) we found experimentally that, within the limits of our resolution, the energy release was the same for either $T_0 = 0.200$ K or $T_0 = 0.205$ K. Thus we assume that this temperature deviation does not influence our results. To test the influence of the initial fast cooldown to this temperature regime we did a reverse experiment, heating up the sample very quickly from T_0 to near T_i , and observing the long-time heat absorption. In spite of the much shortened initial temperature change no substantial deviations from the cooling experiments were found for low $T_i < 1$ K, for which the measuring accuracy was sufficient for this type of experiment. Thus we assume that also the initial cooldown period will have little influence on our results at least for $T_i < 1$ K. Following these considerations, we compare our results with Eq. (3), which predicts the time dependence observed in Fig. 2, and fit a family of curves to our data by a suitable choice of the single free parameter \bar{P} . With $\bar{P} = 3.3 \times 10^{31}$ erg⁻¹ cm⁻³ the broken lines in Fig. 2 are obtained, which describe the experimental results quite well even at higher T_i . Black⁴ found $\bar{P} = 2.1 \times 10^{31}$ erg⁻¹ cm⁻³ by application of the tunneling model to the phonon-scattering experiments of Golding and Graebner.¹² The latter experiments probe fast-relaxing excitations, i.e., quite another range of relaxation times, and the similarity of both values is remarkable. In view of the simplifications made in our discussion of the experiment, and in view of the limitations of the standard tunneling model to describe unambiguously the peculiarities of the amorphous state,⁴ we shall not at the present pursue this point further.

Independent of any model, our measurements show that there exists a considerable density of LEE in Suprasil W with relaxation times between $50 < \tau < 6000$ s. This is in contradiction to the recent results of Lopenon *et al.*,⁷ who find an upper limit of relaxation time of order 10 ms in Suprasil W.

We have also studied a sample⁸ of Suprasil I with an OH content of about 1200 ppm and found essentially the same energy relaxation as in Suprasil W. For thermalization temperatures $T_i < 1$ K, the energy release of Suprasil I was slightly increased; for $T_i > 1$ K, it was slightly decreased compared with Suprasil W. This is in keeping with the 10-s specific heats which behave similarly at these temperatures.¹¹ It seems that OH-associated and intrinsic excitations, which

show different relaxation times in echo experiments,¹³ i.e., at short times, cannot be separated on long time scales.

In addition to the results reported here, each cooldown of our samples from room temperature to low temperatures was accompanied by large thermal relaxation effects.¹⁴ Because of the ill-defined cooling procedure, however, no quantitative results are presented.

In summary, we have observed substantial and reproducible thermal relaxation at low temperatures on a time scale of 10^3 s in vitreous silica. We attribute these effects to a slow relaxation of low-energy excitations in the amorphous state. The method presented seems to be well suited for such investigations. On the basis of the standard tunneling model we have tentatively derived a density of states for these excitations, which is comparable in magnitude to that of the more strongly coupled, faster-relaxing excitations, which contribute to phonon scattering. We have found no upper limit for the relaxation times in contrast to recent experiments on the time dependence of the specific heat.

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Dislocation Motion in hcp ⁴He

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Low-frequency measurements of the shear modulus and internal friction of pure and ³He-doped crystals of hcp ⁴He are reported. Previously unobserved features are seen in both quantities. With use of the parameters obtained from our measurements, it is shown for the first time that all the existing dissipation measurements spanning five decades in frequency are consistent with each other and the Granato-Lücke model of overdamped dislocation motion.

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Dislocation dynamics can be studied by measuring the shear modulus and the internal friction of crystals at different frequencies, impurity levels, and temperatures. Solid ⁴He is nearly an ideal crystal for dislocation studies because of the fact that ³He is the only impurity and can be purified down to 10^{-9} level. Ultrasonic attenuation measurements in hcp ⁴He have shown that only basal-plane ($\perp \hat{c}$ axis) dislocations contribute to

the shear modulus by gliding in the basal plane itself.¹ Recent attenuation measurements as a function of amplitude have demonstrated the pinning of dislocations to ³He impurities at lower temperatures² but the effect is absent above 1.5 K.³ The ultrasound velocity measurements show a dislocation contribution⁴ which is difficult to analyze and whose interpretation is still controversial.⁴⁻⁶ Dissipation measurements in the kilo-