Anomalous Frequency Dependence of the Internal Friction of Vitreous Silica

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(Received 11 November 1999)

The internal friction Q^{-1} and the sound velocity $\delta v/v$ of vitreous silica were measured at very low temperatures using mechanical double paddle resonators operated at frequencies ranging from 0.33 to 14 kHz. Below ~40 mK the internal friction showed an unexpected temperature and frequency dependence, with absolute values of Q^{-1} clearly exceeding those predicted by the standard tunneling model. Even though the most plausible origin of the observed excess internal friction appears to be the mutual interaction between tunneling states, the results are difficult to reconcile quantitatively with present theories taking into account this interaction.

PACS numbers: 62.65.+k, 61.43.–j

At very low temperatures the physical properties of glasses are strongly influenced by the presence of tunneling states [1,2]. These low-energy excitations are formed by small clusters of atoms which can move even at very low temperatures between two almost degenerate configurations by tunneling through the barrier of a double-well potential. Although the microscopic nature of the tunneling states in amorphous solids is not precisely known their influence on the macroscopic quantities of glasses has been quite successfully explained by the phenomenological standard tunneling model (STM) [3]. Aside from the mere existence of double wells this model supposes a broad distribution of the potential parameters—resulting from the large variety of local configurations in a glass—and a coupling between phonons and tunneling states. Below 1 K the dominant interaction process between low-frequency phonons and tunneling systems is assumed to be the one-phonon relaxation [4].

The Tunneling Model provides good overall agreement with experimental findings above ~100 mK for a large variety of glasses. At very low temperatures, however, significant discrepancies occur indicating the relevance of mutual interaction between neighboring tunneling states [5,6] which is not allowed for in the STM. One of the most obvious shortcomings of the STM is that the internal friction of glasses at very low temperatures does not obey the predicted [7] T^3 dependence but exhibits a weaker temperature dependence as was observed in several vibrating reed experiments [8,9]. However, vibrating reeds are known to have fairly large background losses of the order of 10^{-5} or even higher [8]. Hence at low temperatures, when the internal friction of glasses becomes very small, the measured value of Q^{-1} may be strongly influenced or even dominated by the clamping losses, and its temperature dependence may be significantly distorted. Nevertheless it was widely accepted that at temperatures around 10 mK the internal friction of glasses varies approximately linearly with temperature [10–14]. In this Letter we demonstrate that this is not generally the case. Rather, the observed temperature dependence turns out to vary strongly with experimental frequency. A brief account of part of the data has been given elsewhere [15].

The experiments were done using double paddle oscillators of two different sizes laser cut from a 0.4 mm thick plate of vitreous silica [16]. The geometry, very similar to that of silicon oscillators successfully used by the Pohl group [17], is shown in the lower part of Fig. 1. This mechanical resonator can be operated in different torsional (T) and bending (B) modes, i.e., at different frequencies and elastic polarizations. Its major advantage, however, is that only very small strain amplitudes occur at the clamping position. Finite element calculations show that for all modes investigated the strain amplitudes at the clamp are reduced by more than 1 order of magnitude compared to the maximum strain amplitudes

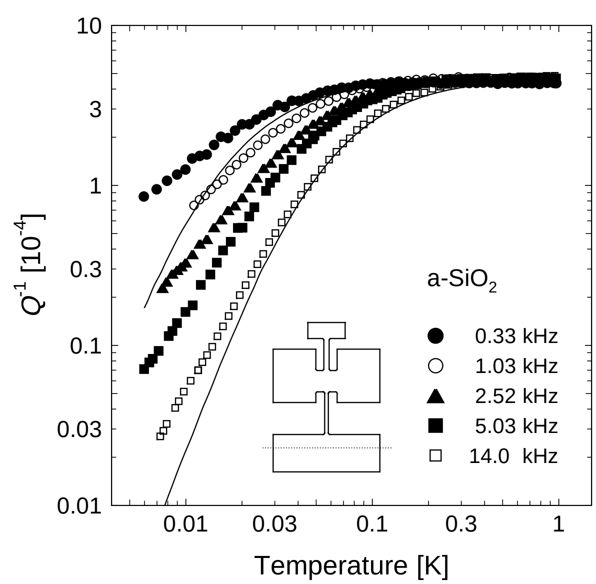

FIG. 1. Temperature dependence of the internal friction of vitreous silica at five frequencies. No background is subtracted. The solid lines are fits according to the tunneling model for frequencies 0.33 and 14 kHz, respectively. Also shown is the geometry of the oscillator, with the thin dotted line marking the clamping position.

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occurring in the sample. Measurements in our group with almost identically shaped silicon oscillators have revealed quality factors Q larger than 10^6 for at least five different modes. Hence we may expect a similarly small background loss of less than 10^{-6} for the glass paddles. The lateral dimensions of the oscillators were 28×20 mm^2 and 16.8×12 mm^2, respectively. Compared to the large oscillator the eigenmode spectrum of the small resonator was shifted by about a factor of 3 to higher frequencies. The large resonator was operated at 0.33 (T), 1.26 (B), 2.52 (B), and 5.03 (T) kHz while the small oscillator was investigated at 0.63 (B), 1.03 (T), 3.88 (B), and 14 (T) kHz. All measurements were done at excitation levels small enough to avoid nonlinear behavior [8,9]; the maximum strain amplitudes in the samples were $\sim 1 \times 10^{-7}$ or smaller. The large (small) sample was covered with a 1.4 (1.1) μm thick silver film. A large film thickness was required to ensure thermalization even at lowest temperatures; however, it was chosen thin enough to have no significant effect on overall damping of the oscillator [18]. Excitation and detection of the oscillator motion was done capacitively [9].

Figure 1 shows the temperature dependence of the internal friction at five frequencies on a double-logarithmic scale. Throughout this paper open and closed symbols denote data obtained from the small and the large paddle, respectively. Above 0.3 K the internal friction is almost independent of temperature and frequency [19]. According to the STM, this plateau value is given by

$$Q^{-1} = \frac{\pi}{2} C . \tag{1}$$

Here $C = \overline{P}\gamma_i^2/\rho v_i^2$ denotes the macroscopic coupling constant, given by the constant density of states of tunneling systems $\overline{P}$, the tunneling system–phonon-coupling γ, the mass density ρ, and the sound velocity v. The index i stands for longitudinal or transversal polarization. Torsional and bending modes show the same absolute value of Q^{-1}; i.e., for vitreous silica the ratio γ_i/v_i does not depend on the elastic polarization—this is in agreement with earlier acoustic measurements [9,20] but has never been demonstrated so clearly by measuring different modes of the same sample.

The most interesting and new observation, however, comes from the analysis of the low-temperature data: In the temperature range 6–30 mK the internal friction of all modes varies in good approximation as $Q^{-1} \propto T^{\alpha}$, with the exponent α increasing monotonically with increasing frequency from values smaller than unity below 1 kHz to values larger than 2 at 14 kHz. The exponents are shown in Fig. 2 as a function of frequency. In this context it is interesting to note that very recent measurements of the internal friction of vitreous silica taken at 90 kHz show, quite consistently with the results of Figs. 1 and 2, an even stronger temperature dependence with an exponent between 2.5 and 3 [21]. However, below 30 mK background losses start to contribute significantly in this experiment and make it difficult to draw unambiguous conclusions on the behavior of Q^{-1} at very low temperatures.

The observed power law behavior is in clear disagreement with the STM that predicts with decreasing temperature a smooth transition from the plateau to a $Q^{-1} \propto T^3$ behavior. It should be noted that, for instance, at 1 kHz the cubic temperature dependence may be expected only at temperatures below $\sim$8 mK; hence the exponents shown in Fig. 2 should not be directly compared to the STM value of 3 valid only at even lower temperatures. More useful is a comparison of the experimental data with numerical calculations according to the tunneling model. Included in Fig. 1 as solid lines are two numerical fits, for the sake of clarity only for the lowest (0.33 kHz) and the highest (14 kHz) experimental frequency, respectively. Two free parameters enter the calculation, namely, the macroscopic coupling constant $C = 2.8 \times 10^{-4}$, which can be easily estimated using Eq. (1), and the prefactor $A = 8 \times 10^7$ K^{-3} s^{-1} of the one-phonon relaxation rate [4]. This value is in good agreement with previous estimates [8,9]. One can see in Fig. 1 that the observed internal friction starts to significantly exceed the calculated values below $\sim$40 mK and that the relative deviation between data and fit becomes larger with decreasing temperature. This becomes even more obvious by plotting the ratio between the experimental data Q^{-1} and the numerical calculation Q^{-1}_{STM}; see Fig. 3. The same parameters C and A were used for all frequencies. Quite remarkably, all curves show even quantitatively the same strong increase towards very low temperatures; i.e., the ratio $Q^{-1}/Q^{-1}_{\mathrm{STM}}$ appears not to depend on frequency.

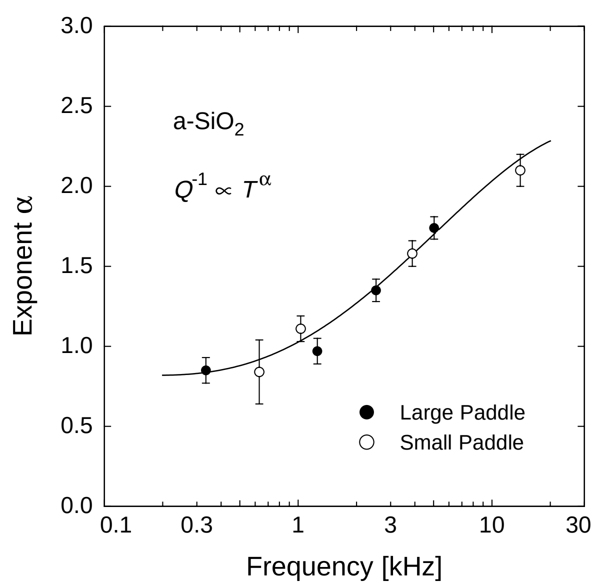

FIG. 2. Exponent α derived from the temperature dependence $Q^{-1} \propto T^{\alpha}$ of the internal friction of vitreous silica below 30 mK. The open and closed symbols denote values obtained from measurements with the small and the large paddle oscillators, respectively.

What is the origin of the observed excess internal friction? There is strong evidence from a large variety of experiments on glasses that the mutual interaction of tunneling states is of great importance at temperatures well below 100 mK [5,6,22], and we suppose that interaction effects are responsible for our observations, too. Several approaches taking into account this interaction have already been suggested to explain the deviation from the T^3 behavior of the internal friction. Burin and Kagan [10,11] proposed the occurrence of pair excitations due to strain-mediated interaction between tunneling states. The interaction between pairs with similar tunnel splitting leads to an additional relaxation contribution with a rate [11]

$$\tau_{\mathrm{p}}^{-1} \simeq \frac{10 k_{\mathrm{B}} C^3}{\hbar} \left(\frac{\Delta_0}{E}\right)^2 T\,. \tag{2}$$

Here Δ_0 denotes the tunnel splitting and E denotes the energy splitting of the two-level systems. Note that no additional free parameters are introduced in Eq. (2). As a consequence of Eq. (2), a linear temperature dependence of the internal friction is expected at very low temperatures. We have tried to incorporate the relaxation rate (2) in our numerical calculations assuming that the total relaxation rate is given by the sum of τ_{p}^{-1} and the one-phonon rate. However, no agreement was obtained because in the temperature range of our experiments Eq. (2) gives only a negligibly small contribution. In fact, the crossover from the one-phonon dominated relaxation to the linear temperature dependence is expected to occur at a temperature $T^\star \simeq (10 k_{\mathrm{B}} C^3/\hbar A)^{1/2}$ [11]. Putting in the values of C and A we obtain $T^\star \simeq 0.6$ mK [23]. It turns out that the numerical prefactor of Eq. (2) would have to be increased arbitrarily by almost 4 orders of magnitude to achieve good agreement with the internal friction data. While it is interesting to note that an additional relaxation contribution of the form $\tau^{-1} \propto (\Delta_0/E)^2 T$ does in principle describe the data fairly well, the Burin model appears not to be applicable unless a theoretical justification for the required enormous enhancement of the prefactor can be given.

A somewhat different approach to take into account interaction between tunneling states was given by two of us [6]. Based on a theory worked out by Würger for the tunneling of substitutional defects in alkali halides [24,25] a very simplified picture for the possible influence of interaction between tunneling states in glasses was developed. In this model the interaction leads to an incoherence of the tunneling motion at very low temperatures. As a result, additional relaxation effects occur and modify the temperature dependence of $\delta v/v$ and Q^{-1}. Moreover, the resonant contribution to $\delta v/v$ is expected to be significantly reduced [6]. While the basic idea of this model appears to be quite promising a fully developed theory for incoherent tunneling in glasses at very low temperatures has not been worked out yet, and therefore a sound analysis of our data seems to be hardly possible at present within this approach.

Finally, we show in Fig. 4 the temperature dependence of the sound velocity $\delta v/v$ at four frequencies. For all modes the sound velocity increases at low temperatures, passes a maximum, and decreases at higher temperatures where one-phonon relaxation contributes significantly. As in previous experiments on glasses [9,20,26] the increase of the sound velocity below the maximum has a similar slope as the decrease above the maximum, in contrast to the expectation of the STM where a slope ratio of $2:(-1)$ is predicted. While the principal behavior has been reported before and tentatively been explained by incoherent tunneling [6] we want to point out a new observation: The low-temperature slope of the sound velocity

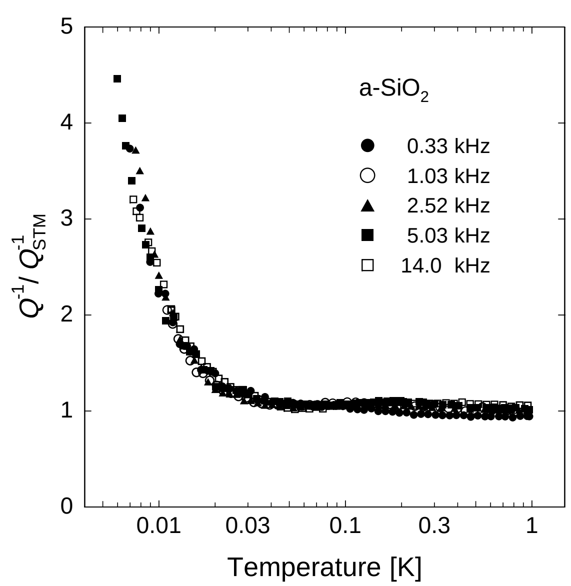

FIG. 3. Experimental data of Fig. 1 divided by numerical fits according to the standard tunneling model. The parameters used for the numerical calculations are discussed in the text.

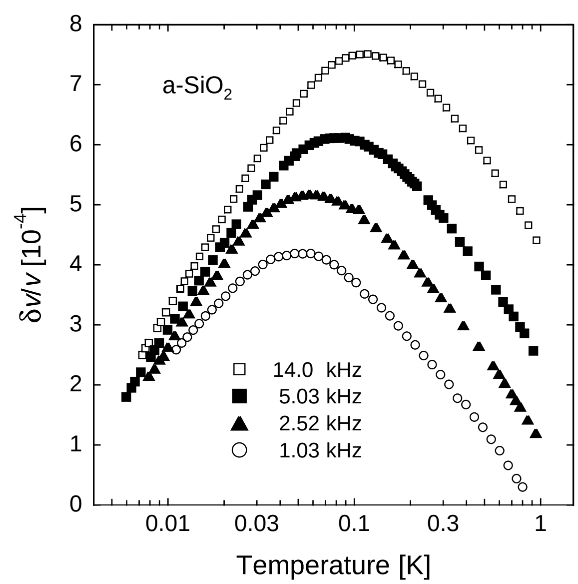

FIG. 4. Temperature dependence of the sound velocity of vitreous silica at four frequencies.

appears to slightly vary with experimental frequency. As the low-temperature part of $\delta v/v$ is almost entirely determined by the resonant interaction between phonons and tunneling states this frequency dependence indicates a frequency dependence of the resonant interaction. Additional measurements over a wider range of frequencies and temperatures are clearly desirable to establish this interesting observation.

The data on the acoustic properties of vitreous silica presented here are a further proof that the simple picture of noninteracting tunneling states is not sufficient at temperatures well below 100 mK. It should be emphasized that minor modifications of the STM such as changes of the distribution function of the double-well parameters cannot resolve the discrepancies between theory and experiment. The results rather indicate that fundamental revisions of our understanding of the dynamic properties of glasses at very low temperatures are necessary. A new approach to consider interaction effects of tunneling states has been suggested very recently [27] to explain the unexpected magnetic field dependence of the dielectric properties of multicomponent glasses [22]. In this theory a quasiparticle picture was introduced for the low-energy excitations in glasses at very low temperatures. It appears very promising to investigate if this model is applicable not only to the dielectric but also to the elastic properties of glasses. Measurements of the sound velocity and internal friction in magnetic fields using the very accurate double paddle oscillator technique may yield interesting answers to this question.

We thank S. Hohmann for support with the experiments. Valuable discussions with R. O. Pohl, A. Burin, and I. Polyshchuk are gratefully acknowledged. We are also grateful to S. Wächter from IFW Jena, Germany, for the high-quality laser cuts of the samples. This work was supported by the Deutsche Forschungsgemeinschaft (Grant No. Hu359/11).

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