Acoustic Properties of Amorphous Silica between 1 and 500 mK

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We have made reliable measurements of the sound velocity $\delta v/v_0$ and internal friction Q^{-1} in vitreous silica at 1.03, 3.74, and 14.0 kHz between 1 mK and 0.5 K. In contrast with earlier studies that did not span as wide a temperature and frequency range, our measurements of Q^{-1} reveal a crossover (as T decreases) only near 10 mK from the T^3 dependence predicted by the standard tunneling model to a T dependence predicted if interactions are accounted for. We find good fits at all frequencies using a *single* interaction parameter, the prefactor of the interaction-driven relaxation rate, in contrast to earlier claims of a frequency dependent power law. We also show that the discrepancy in the slopes $d(\delta v/v_0)/d(\log_{10}T)$ below and above the sound velocity maximum (1:−1 observed, 1:−2 predicted) can be resolved by assuming a modified distribution of tunneling states.

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Much of glassy low temperature behavior, first observed in [1], can be understood in terms of the standard tunneling model (STM) [2–7], but this model has some inadequacies [8,9]. In particular, the mechanisms responsible for dissipation in the presence of the disorder and low phonon density that characterize amorphous solids at very low temperatures have been a matter of recent debate. The internal friction Q^{-1} was measured in the present work in order to shed light on this question.

Several theories have been proposed in response to observations that could not be explained within the STM. These include explanations for the quantitative universality of glass [10–13]. In specific glasses, the unexpected magnetic field dependence of the dielectric susceptibility [14–16] and polarization echo amplitude [17] was related to nuclear [18,19] and electronic [20] magnetic and quadrupolar moments of two level systems (TLS). Furthermore, it was shown that the nuclear quadrupole interaction could be responsible for the saturation of the dielectric constant observed in several glasses below 5 mK [21]. Directly related to the present work is theoretical work on the effects of interacting pairs of TLS in any glass: nonequilibrium dielectric and acoustic measurements reported in [22–24] were explained in terms of a dipole gap [25]. A model of interacting pairs [26] was also used to explain measurements of *equilibrium* dielectric loss [23] in excess of the STM prediction, but only partial agreement between experiment and theory was obtained. Equilibrium acoustic measurements in disagreement with the STM have also been reported in several works [27–31]. In particular, in [29] internal friction (Q^{-1}) measurements were interpreted in terms of interacting pairs [26] and incoherent tunneling [32], but a full agreement with theory was not obtained.

In this Letter, we show that our results do not agree with the STM, but that they are also different from the results in [29]. We show that all of our low temperature data can be fitted by adding a single additional parameter to the STM, i.e., an interaction-driven relaxation rate $1.0\times 10^5 T\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$ [26], suggesting that interactions lead to dissipation at the lowest temperatures. By assuming an atypical TLS distribution function, we also fit the theory to the $\approx 1{:}-1$ ratio of $d(\delta v/v_0)/d(\log_{10}T)$ below and above the sound velocity maximum exhibited by our data. A brief account of part of the sound velocity data has been given elsewhere [33,34].

The acoustic properties of glass in the absence of interactions between tunneling states can be understood in terms of the STM [7], in which tunneling states are approximated by TLS with a distribution $P(\Delta, \Delta_0) = P_0/\Delta_0$ of asymmetries Δ and tunneling amplitudes Δ_0 and an energy splitting $E = \sqrt{\Delta^2 + \Delta_0^2}$. For $k_B T/\hbar\omega \gg 1$, as in the present work, resonant processes cause a change in sound velocity $\delta v/v_0$ relative to the sound velocity of the host $v_0 \equiv v(T = 0)$ but cause negligible internal friction Q^{-1}. Additionally, phonons perturb the asymmetry Δ of the TLS, resulting in a nonequilibrium distribution of tunneling state occupancies and subsequent relaxation toward the equilibrium state. Such relaxation contributes to $\delta v/v_0$ and Q^{-1}, and its contribution to Q^{-1} drops off as T^3 at low T. Motivated by [26,35], we will add to the phonon-driven relaxation rate an interaction-driven rate $\gamma_{\mathrm{tr},m} = bT$, where b is a constant, which leads to $Q^{-1} \propto T$ at low T. See [36] for a detailed derivation of the tunneling model predictions.

The experiment was carried out by electrostatically driving and detecting the motion of an amorphous silica oscillator with a double paddle geometry nearly identical to the one described in [29]. The oscillator has lateral dimensions of ≈ 1 cm and a thickness of 0.4 mm with resonant modes in the kilohertz frequency range. The geometry of the oscillator as well as the displacement and strain energy density profiles resulting from a finite element calculation are shown in the inset of Fig. 1. For

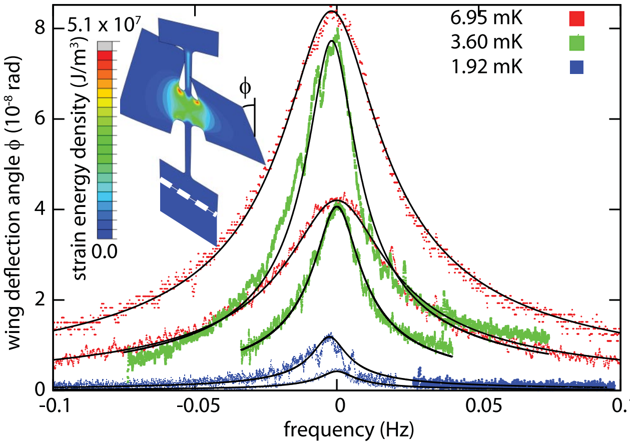

FIG. 1 (color). Response of the 14.0 kHz mode at three different temperatures. For each temperature two drive levels were chosen to demonstrate the linear response. Inset: Finite element visualization of the oscillator displacement and strain energy density (linear scale) at 14.0 kHz for 0.4 rad peak displacement. The white broken line indicates the upper edge of the dry clamp.

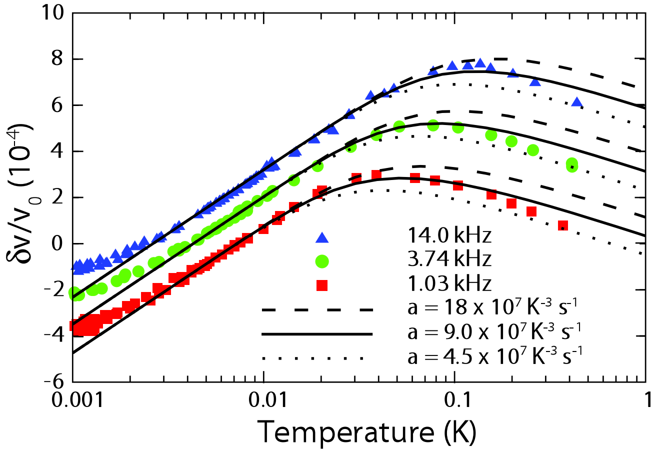

FIG. 2 (color online). Measurements of the relative change in sound velocity $\delta v/v_0$ (data points) and predictions of the STM [$b = 0$] (curves) for each of the experimental frequencies (offset vertically). The curves correspond to a tunneling strength $C = 2.4 \times 10^{-4}$ and the indicated values of a, the prefactor of the single-phonon relaxation rate. The best fit at each frequency is represented by a solid curve. These curves are indistinguishable from those corresponding to the same C and a, but with the prefactor $b = 1.0 \times 10^5\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$ (see text).

clarity, the displacement profile shown here is for 0.4 rad peak displacement; the paddle was in fact operated at very low amplitudes in order to remain in the linear regime. The displacement near the clamp is very low for this mode, resulting in a very small clamping loss contribution to Q^{-1}. The surface of the paddle was coated with a micron-thick silver film for thermalization. Measurements of an equivalent film on a single crystal silicon substrate (details to be published elsewhere) revealed a temperature independent contribution to the dissipation $\Delta Q^{-1} = 3 \times 10^{-7}$ below 10 mK, which was subtracted from the data.

A sample of the raw data at 14.0 kHz for two different drive levels at each of three temperatures is shown in Fig. 1. The center frequency for data taken at low drive for each temperature was shifted to zero frequency, but any frequency offset between the high and low drive data at a given temperature was retained. It was important to lower the drive level as the temperature decreased in order to remain in the linear regime, where the data exhibit the Lorentzian form shown by the black lines in the figure. The frequency shift between low and high drive data sets at each temperature is negligible on the scale of the variation of $\delta v/v_0$ with temperature and is accounted for by the small thermal drift of the low temperature stage. Furthermore, the results of an ABAQUS finite element method calculation imply that the theoretical condition of linearity [37], $\epsilon g/k_B T \ll 1$, where ϵ is the peak strain of the paddle and g is the deformation potential, is satisfied for the measurements in Fig. 1. Care was taken to prevent experimental artifacts at other T and ω as well.

We used the low strain measurements in Fig. 1 and those at other T and ω to obtain $\delta v/v_0 \approx (f_r - f_{r,0})/f_{r,0}$ (assuming negligible thermal expansion) and $Q^{-1} = \Delta f/f_r$, where f_r is the frequency at peak response and Δf is the half-power width. Ideally $f_{r,0} = f_r(T = 0)$, and any small difference amounts to an offset in $\delta v/v_0$, which is not of interest here. These results are shown along with the predictions of the STM in Figs. 2 and 3. The solid curves in Fig. 2 and the red and blue curves in Fig. 3 correspond to the STM with best fit values of the tunneling strength $C = 2.4 \times 10^{-4}$ and prefactor of the single-phonon relaxation

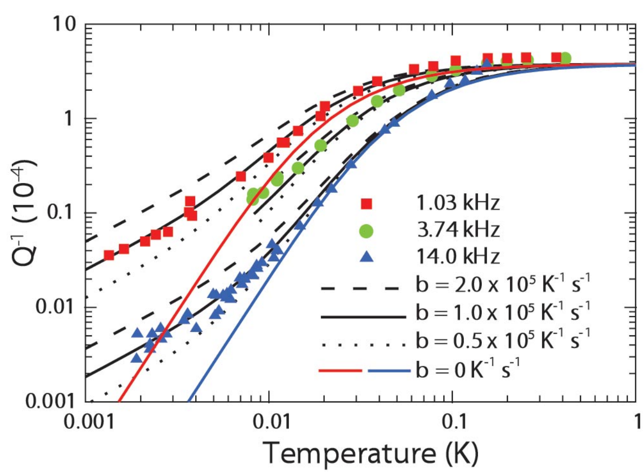

FIG. 3 (color). Measured internal friction Q^{-1} (data points) and predictions (curves) for each of the experimental frequencies. The curves correspond to a tunneling strength $C = 2.4 \times 10^{-4}$, a single-phonon relaxation prefactor $a = 9.0 \times 10^7\ \mathrm{K}^{-3}\,\mathrm{s}^{-1}$, and various values of b. The solid black curves correspond to the best fit value of b and agree well with the measured Q^{-1}. For comparison, the prediction of the noninteracting model ($b = 0$) is shown for $\omega/2\pi = 1.03$ and 14.0 kHz.

rate $a = 9.0 \times 10^7\ \mathrm{K}^{-3}\,\mathrm{s}^{-1}$, and the dashed and dotted curves in Fig. 2 show the sensitivity of the fit to the parameter a. The best fit values obtained here are comparable to those in [29].

The discrepancy between the measurements of Q^{-1} and the predictions of the STM below 10 mK shows most clearly that the present results are not in agreement with the STM. Furthermore, the ratio of $d(\delta v/v_0)/d(\log_{10}T)$ at low and high temperatures is more nearly $1{:}-1$ than the $2{:}-1$ ratio predicted by the STM, and $\delta v/v_0$ departs from the $\log T$ dependence at $T < 3$ mK. The latter effect could be due to thermal decoupling of the sample from the experimental plate, but this is unlikely because of the relatively large amount of heat required to produce the flattening [34]. Rather, the leveling off may be due to an unknown mechanism intrinsic to the glass. Whatever the origin, it must be distinct from the explanation for the departure of Q^{-1} from the STM prediction since the latter begins at a higher temperature, i.e., 10 mK.

Figure 3 shows the internal friction data from the present experiment along with predictions including the relaxation rate bT. The best fit values of C and a were retained and the new interaction parameter b was varied. The best fit (solid black lines) corresponds to a *single* value of $b = 1.0 \times 10^5\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$. Curves corresponding to a factor of 2 variation in b are also drawn to show the sensitivity to the choice of b. For comparison with the STM prediction, curves corresponding to $b = 0$ and $\omega/2\pi = 1.03$ and 14.0 kHz are also shown. The result with $b > 0$ for the sound velocity is not shown because for $a = 9.0 \times 10^7\ \mathrm{K}^{-3}\,\mathrm{s}^{-1}$ and $b = 1.0 \times 10^5\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$ it is indistinguishable from the STM prediction (Fig. 2). Thus the most striking discrepancy between the STM and the present data, i.e., the behavior of Q^{-1} for $T < 10$ mK, is resolved by assuming the relaxation rate bT. Furthermore, the quality of the fit to Q^{-1} for $T > 10$ mK and to $\delta v/v_0$ over the entire temperature range is maintained or slightly improved.

The origin of the additional relaxation rate assumed above may be interactions between pairs of TLS. It is argued in [26] that the existence of such four-particle clusters leads to a relaxation rate that is linear in temperature. As noted in [35], the prefactor of the term that is linear in temperature must be determined by experiment at this point. While [38] also predicts a relaxation rate that is linear in temperature, according to [35,39] the number of resonant triples of TLS was overestimated.

There are significant differences between the present work and [29], which did not extend to as low a temperature. In [29], the temperature dependence of Q^{-1} was characterized by the power laws T^{α}, which did not have a direct connection to theory, and a monotonic frequency dependence of α was noted. While our data could in principle be so fitted with different values of α over the same limited ($30 > T > 6$ mK) temperature range, it became evident after pushing to lower T that the exponents α relate to the crossover from the developing T^3 STM behavior to the linear temperature dependence generated by interaction-driven relaxation (see Fig. 3). Thus we have shown that it is not necessary to assume a range of power laws to fit the data. We also note that all Q^{-1} in [29] are greater than those observed in our experiment (and differ significantly from those reported in [40]). Third, the frequency dependence of the slope $d(\delta v/v_0)/d(\log_{10}T)$ observed in [29] was not observed in the present work. Thus, the extension of the $\delta v/v_0$ measurements to a lower temperature in the present work allowed for a more definitive measurement of $d(\delta v/v_0)/d(\log_{10}T)$ at 1.0 kHz than in [29].

By assuming a modified TLS distribution function $P_0(1-r)^{\mu-1/2}/2r$ [41], where $r = (\Delta_0/E)^2$, we were able to fit the $\approx 1{:}-1$ slope ratio exhibited by our $\delta v/v_0$ data. See [36] for details. For $\mu = 0$, this distribution reduces to that in [2], which was used for the fits in Figs. 2 and 3. Our $\delta v/v_0$ and Q^{-1} data at 1.03 kHz are shown in Fig. 4 along with the calculations assuming $\mu = 0.09$, $C = 2.9 \times 10^{-4}$, $a = 6.0 \times 10^7\ \mathrm{K}^{-3}\,\mathrm{s}^{-1}$, and $b = 1.0 \times 10^5\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$. Similar agreement between theory and data was obtained at 3.74 and 14.0 kHz using the same fitting parameters. The fit of the STM to thermal properties of glass is not significantly degraded by assuming $\mu = 0.09$: the heat capacity remains nearly linear in T and the thermal conductivity $\kappa \propto T^2$ exactly [41]. While we have no physical justification for this particular distribution, we have shown that it is possible to account for the behavior of Q^{-1} and $\delta v/v_0$ independently from TLS interactions for $T > T_\omega$, the temperature at which the maximum in $\delta v/v_0$ occurs, i.e., at which $\omega = \gamma_{\mathrm{ph},m}(E = k_B T)$.

To conclude, after taking care to eliminate experimental artifacts such as nonlinearity from our measurements, we observed a departure of Q^{-1} from the prediction of the STM below 10 mK and a $\approx 1{:}-1$ ratio for the slopes

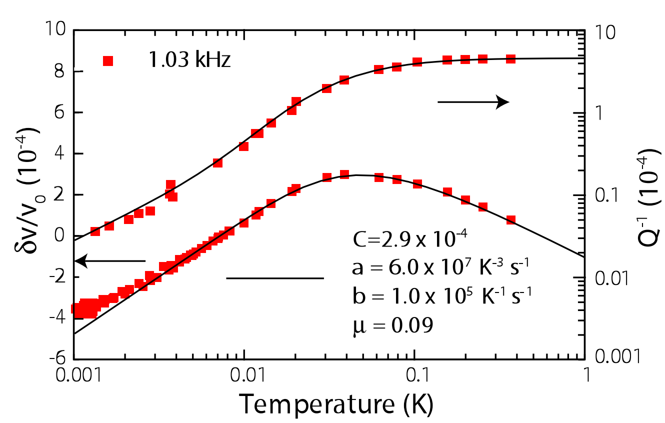

FIG. 4 (color online). Measured $\delta v/v_0$ (offset vertically) and Q^{-1} at 1.03 kHz along with tunneling model predictions assuming an interaction-driven relaxation rate $b = 1.0 \times 10^5\ \mathrm{K}^{-1}\,\mathrm{s}^{-1}$ and TLS distribution parameter $\mu = 0.09$.

$d(\delta v/v_0)/d(\log_{10}T)$. While the slope ratio could be accounted for by a modification of the TLS distribution function, to our knowledge there is no reasonable modification that can account for the observed Q^{-1}. However, the observed Q^{-1} could be accounted for at all three frequencies by adding a single term, $\gamma_{\text{tr},m} = 1.0 \times 10^5 T\ \text{s}^{-1}\,\text{K}^{-1}$, to the STM relaxation rate, motivated by theoretical work on interacting pairs of TLS [26]. While the theory of interacting pairs cannot predict the absolute value of the interaction-driven relaxation rate, it is argued in [35] that the prefactor needed to fit the present results is reasonable within that framework. Including the additional relaxation term did not degrade the quality of the fit to the Q^{-1} data above 10 mK, nor the fit to the $\delta v/v_0$ data over the entire temperature range. We conclude that the data are well described by the addition of a relaxation rate that is linear in temperature, but that the exact origin of the additional relaxation remains as a matter of theoretical debate. Thus, we have presented evidence that a refinement of the widely accepted standard tunneling model for glasses to account for interactions is necessary to describe the acoustic behavior of the prototypical glass, SiO_2, only below ≈ 15 mK.

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