Glass Anomaly in the Shear Modulus of Solid ^{4}He

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The shear modulus of solid H^{4}e exhibits an anomalous increase at low temperatures that behaves qualitatively similar to the frequency change in torsional oscillator experiments. We propose that this stiffening of the shear modulus with decreasing temperature can be described with a glass susceptibility assuming a temperature-dependent relaxation time $\tau(T)$. Below a characteristic crossover temperature T_X, where $\omega\tau(T_X) \sim 1$, a significant slowing down of dynamics leads to an increase in the shear modulus. We predict that the maximum change of the amplitude of the shear modulus and the height of the dissipation peak are independent of the applied frequency ω. Our calculations also show a qualitative difference in behavior of the shear modulus depending on the temperature dependence of $\tau(T)$

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The low-temperature anomaly of solid helium in torsional oscillators (TO) reported by Kim and Chan [1] has inspired an intense search for mechanical and thermodynamic anomalous properties. The observed increase of the resonance frequency below ~200 mK was taken as evidence for mass decoupling due to supersolidity—a quantum solid that can sustain mass superflow without dissipation. It is now accepted that the presence of defects in solid ^{4}He is required to produce supersolid signatures. In addition, it has been speculated that supersolidity may occur along dislocation lines [2,3] or grain boundaries [4–6] in solid helium. Direct experimental evidence for a true phase transition into a supersolid state remains inconclusive. To date no definitive sign of Bose-Einstein condensation (BEC) has been seen in measurements of the mass flow [5,7,8], the melting curve [9], and the lattice structure [10,11].

The basic issue with the unambiguous identification of the BEC features arises from the presence of defects that display their own dynamics and contribute to observables in the same temperature and pressure range, where supersolid anomalies are expected. We therefore are left with a dilemma: on one hand, defects are required to produce supersolidity; on the other hand, defects exhibit their own dynamics. Thus, any unambiguous identification of a possible supersolid state relies on a detailed understanding of the behavior of defects. For that reason we consider a theoretical framework that captures the dynamics of defects in solid ^{4}He in the form of a glassy component, which makes up a small fraction of the crystal [12,13]. This glassy component is suggested to cause the TO and thermodynamic anomalies. Further, it is consistent with reported signatures of long equilibration times, hysteresis, and a strong dependence on growth history. The glass may be created from distributions of crystal defects forming two-level-systems (TLS) [14,15]. To determine the nature of the TLS, a detailed microscopic characterization of samples is needed. Possible candidates for the microscopic realization of the TLS are *pinned* segments of dislocation lines [12,13,16], which naturally occur in bulk and confined solid helium. These defects can be annealed away and hence drastically change the mechanical properties of the solid. We demonstrated in previous work [12,13,16–18] that the freezing out of excitations can account for the anomalies in TO and in thermodynamic experiments; a relaxation time that increases with decreasing temperature is required to describe the low-temperature anomalous features.

Here we consider the dynamic response of elastic properties in ^{4}He crystals [19–25]. Very recent shear modulus measurements [19–21] reveal qualitative similarities with the TO experiments [1,26–32]. In the shear modulus experiment, the solid helium is grown in between two closely spaced sound transducers. When one of the transducers applies an external strain, the other transducer measures the induced stress from which the shear modulus of the sample is deduced. The experiment thus provides a direct measurement of the elastic response to the applied force within a broad and tunable frequency range. The frequency dependence is especially crucial in determining the nature of the relaxation processes, which is the main focus of this Letter.

We analyze the shear modulus within the glass framework where the amplitude of the shear modulus increases (stiffens) upon lowering T because of excitations freezing out. This is accompanied by a prominent dissipation peak, indicative of anelastic relaxation processes. These anomalies happen in the same temperature range where the TO anomalies were found. By studying the glass model for a complex shear modulus $\mu(\omega; T)$ we find the following. (1) The damping and amplitude of vibrations in ^{4}He are controlled by freeze-out dynamics. They occur at temperatures where $\omega\tau(T) \sim 1$. In our picture the glassy contribution represents a small fraction of the total response because the glass occupies only a small fraction of the solid. Thus we propose a theoretical description of an elastic material with a small anelastic component that is modeled by a glassy susceptibility. (2) We calculate the amplitude of the shear modulus in agreement with shear mode experiments. Within this approach we find that the maximum of the shear modulus change and the height of the dissipation peak are independent of frequency. (3) From our model independent susceptibility analysis of the Cole-Cole plot, we derive a universal parameter that can describe either thermal or nonthermal activation. (4) We predict a relation for the inverse crossover temperature $1/T_X$ versus the applied frequency ω; i.e., $\omega\tau(T_X) = 1$.

Model.—We investigate the dynamics of the solid using linear response theory with a backaction term [13,33]:

$$\rho\partial_t^2 u_i + \partial_j \sigma_{ij}^{\mathrm{He}} = f_i^{\mathrm{EXT}} + f_i^{\mathrm{BA}}, \tag{1}$$

where ρ is the mass density and u_i is the displacement in the ith direction. f_i^{EXT} and f_i^{BA} are the external force density and the backaction force density in the ith direction. $\sigma_{ij}^{\mathrm{He}}$ is the elastic stress tensor due to solid helium. In general, $\sigma_{ij}^{\mathrm{He}} = \lambda_{ijkl}u_{kl}$, with the elastic modulus tensor λ_{ijkl} [33]. The backaction describes the delayed restoring force of a glass component that backacts on the solid matrix and thus modifies the net force. For simplicity, we consider a homogeneous solid and set the shear wave propagation along the z axis and assume that the wave polarization lies in the x-y plane. For such a case the backaction is

$$f_i^{\mathrm{BA}} = \int_{-\infty}^{t} dt' \mathcal{G}(t, t'; T)\partial_z^2 u_i(t'), \tag{2}$$

where $\mathcal{G}$ describes the strength of the backaction on solid ^{4}He and $i = x, y$. Although f_i^{BA} is typically much smaller than the elastic restoring force $\partial_j \sigma_{ij}^{\mathrm{He}}$, it is in fact this term that is responsible for the anomaly. The isotropic approximation is appropriate for measured polycrystalline and amorphous materials.

Applying the same approximation to the elastic modulus tensor, $\lambda_{ijkl} = \lambda_0\delta_{ij}\delta_{kl} + \mu_0(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$, the elastic stress tensor in Eq. (1) is finite only for orientations $j = z$ and either k or l equal to z. With k, l being interchangeable, the relevant element will be λ_{iziz}, which gives μ_0. The fully dressed shear modulus relates the displacement to an external force, or $\rho\partial_t^2 u_i + \mu\partial_z^2 u_i = f_i^{\mathrm{EXT}}$. Comparing this expression with Eq. (1) we obtain

$$\mu(\omega; T) = \mu_0(T) - \mathcal{G}(\omega; T). \tag{3}$$

The backaction may be described by a distribution of Debye relaxors $\mathcal{G} = \int_0^\infty dt P(t)[1 - 1/(1 - i\omega\tau t)]$, with the dimensionless parameter t, the normalized distribution of relaxation times $P(t)$, and the relaxation time of the glass τ. In general, $\tau(T)$ increases with decreasing T and approaches infinity at the ideal glass temperature T_g. The specific form of $\tau(T)$ can change qualitatively the T dependence of μ and will be discussed. For simplicity, we choose for $\mathcal{G}$ the Cole-Cole distribution. Integrating over $P(t)$ yields [17]:

$$\mathcal{G}(\omega; T) = g_0/(1 - (i\omega\tau)^\alpha), \tag{4}$$

$$\mu(\omega; T) = \mu_0\{1 - g/[1 - (i\omega\tau)^\alpha]\}, \tag{5}$$

with the renormalized parameter $g \equiv g_0/\mu_0$, which is sample dependent. The experimental measurables are the amplitude of the shear modulus $|\mu|$ and the phase delay between the input and readout signal, $\phi \equiv \arg(\mu)$; ϕ measures the dissipation of the system, which is related to the inverse of the quality factor $Q^{-1} \equiv \tan\phi$.

Several interesting results follow from Eq. (4). First, the change in shear modulus $\Delta\mu$ between zero and infinite relaxation time is $\Delta\mu/\mu_0 = g$, measuring the strength of the backaction as well as the concentration of the TLS. Second, the peak height $\Delta\phi$ of the phase angle is proportional to g. When $\omega\tau = 1$ (location of dissipation peak), $\Delta\phi$ becomes approximately $\Delta\phi \approx g\cot(\alpha\pi/4)/(2 - g)$ when $g \ll 1$. For $1 < \alpha \leq 2$, this simplifies even further:

$$\Delta\phi \approx (1 - \alpha/2)(\Delta\mu/\mu_0), \tag{6}$$

where $\Delta\phi$ is in units of radian. The peak height $\Delta\phi$ depends only on the phenomenological glass parameters α and g, so both $\Delta\mu$ and $\Delta\phi$ are independent of frequency.

Finally, we comment that a glass exhibits viscoelastic properties [34]. So far the response formulation is equivalent to a viscoelastic system with a Cole-Cole distribution of relaxation processes also known as a generalized Maxwell model—parallel connections of an infinite set of Debye relaxors each with a different τ.

Results.—We now compare our glass model calculations with the experimental shear modulus measurements by Syshchenko and co-workers [21] for a transducer driven at 200 Hz (SM200), 20 Hz (SM20), and 2 Hz (SM2) [35].

To model specific examples of the T-dependent $\tau(T)$ in Eq. (4), we consider Vogel-Fulcher-Tammann (VFT) and power-law (PL) relaxation processes, which represent thermal and nonthermal activation processes, respectively. For the VFT we assume the general form:

$$\tau(T) = \begin{cases} \tau_0 e^{\Delta/(T-T_g)} & \text{for } T > T_g \\ \infty & \text{for } T \leq T_g. \end{cases} \tag{7}$$

Here τ_0 is the attempt time and Δ is the activation energy. The experimental data for all three frequencies are described by a single set of model parameters shown in Fig. 1. In our parameter search we did not bias T_g to be positive (real transition at T_g, where τ diverges) or negative T_g. Good agreement between calculations and experiments is obtained for all three frequencies for $T_g = -30$ mK. We refer to this calculation as "VFT$_<$." Note that the physical meaning of a negative T_g is that no real phase transition occurs. The origin of a negative T_g may be an inherent quantum fluctuation phenomenon due to the large zero-point motion of helium, which suppresses the onset of a lower phase transition. This may be analogous to a negative Curie-Weiss temperature in magnetism in the presence of antiferromagnetic fluctuations.

We calculated for $VFT_<$ the shear modulus versus temperature to compare with experiment and obtained that both amplitude and phase angle agree well with experiment; see Fig. 1. Our calculations confirm that the change of amplitude and peak height of phase angle is nearly frequency independent. By defining the crossover temperature T_X as the point where the phase angle exhibits a peak corresponds for the Cole-Cole distribution to a turning point in the amplitude. As expected, we find that T_X decreases with decreasing ω. Since the phase angle measures dissipation, it can provide valuable information about the underlying relaxation processes. For example, additional dissipation seen at higher frequencies may be caused by additional dissipation mechanisms, e.g., dislocation or vacancy motion, not included in this theory. Finally, when we set $T_g = 0$ K the VFT expression reduces to an Arrhenius rule "VFT_0." While it can describe T_X reasonably well, the Arrhenius relaxation shows a linewidth much narrower than $VFT_<$ (not shown), which is not in accord with the data.

Next we show in Fig. 2 the Cole-Cole plots for experiments and calculations. The experimental Cole-Cole curves for different frequencies collapse roughly onto one curve confirming our theoretical assumption that $\omega\tau(T)$ is a universal parameter [36]. In addition, the data show mirror symmetry about $\mathrm{Re}[|\mu - \mu_0|/\Delta\mu] = 0.5$ in support of the Cole-Cole distribution used. The discrepancy between theoretical and experimental data may be explained by additional relaxation processes occurring at temperatures above T_X, consistent with the excess dissipation seen at higher frequencies. Finally, the scaling of the Cole-Cole plot with $\omega\tau(T)$ is a general feature of glasses that is applicable beyond shear modulus experiments. Indeed, it has also been seen in the TO experiments [31].

To search for a possible phase transition, we study the higher and lower frequency behavior for various relaxation scenarios. For the Cole-Cole distribution the crossover happens when $\omega\tau(T) = 1$. From that we estimate T_X as a function of the applied frequency $f = \omega/2\pi$. Figure 3 shows $1/T_X$ versus f. The $VFT_<$ and VFT_0 calculations give a significantly better description than the PL calculations. The $VFT_<$ line is convex, while the Arrhenius line VFT_0 is straight. The upward curvature is typical for a VFT relaxation time with $T_g < 0$, as opposed to $T_g > 0$. For comparison, the power-law predictions are also shown for phase transitions occurring at 0 K (PL_0) and 40 mK (PL_{40}). For positive T_g (see PL_{40}), there is a true freeze-out transition, indicating the arrested dynamics for $f \to 0$ Hz. For both VFT and PL relaxation times our calculations demonstrate that in the low frequency limit the existence of a phase transition should show clear signatures of T_X converging toward the ideal glass temperature T_g. This behavior can serve as experimental evidence for a possible phase transition.

In summary, we have shown that the low-temperature shear modulus anomaly of solid ^{4}He can be described using the theoretical framework of glasses for which we predict characteristic dissipation signatures. The elastic shear modulus is strongly affected by the dynamics of defects. The freezing out of excitations leads to a stiffening

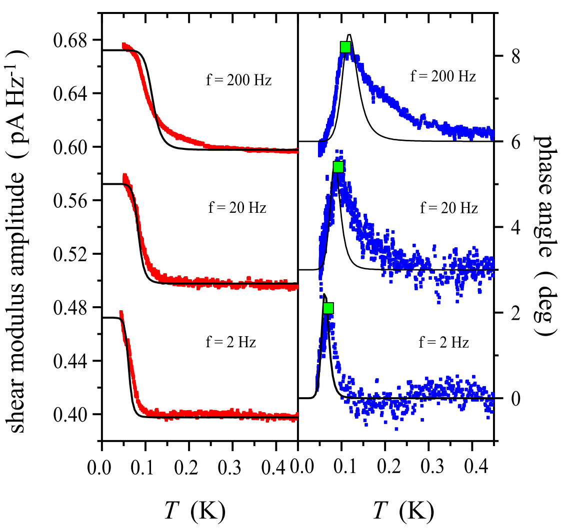

FIG. 1 (color online). Experimental data and calculations (VFT) of the shear modulus versus temperature. The square symbols are the experimental data for the shear modulus amplitude and the phase angle. The black lines are the results from calculation. The big green squares mark the peak location. The calculation uses $\alpha = 1.31$, $g = 1.44 \times 10^{-1}$, $\mu_0 = 0.47$ pA Hz^{-1}, $\tau_0 = 50$ ns, $\Delta = 1.92$ K, and $T_g = -69.3$ mK. The shear modulus amplitude and phase angle plotted is shifted by 0.1 pA Hz^{-1} and 3° with respect to the 2 Hz data.

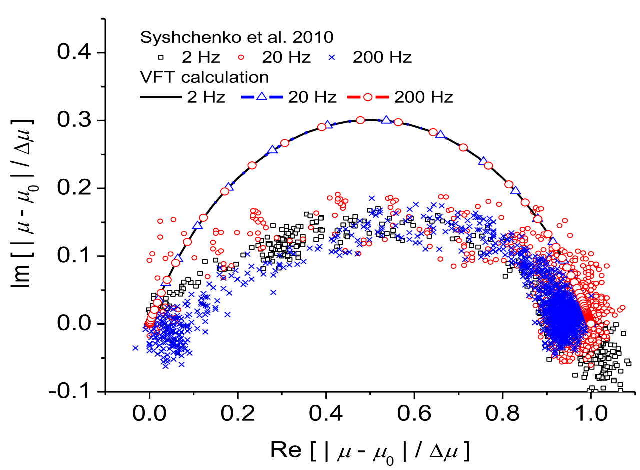

FIG. 2 (color online). The Cole-Cole plots for experimental data and for $VFT_<$ (see text). For given form of τ, all different frequency curves collapse onto one single master curve reflecting that $\omega\tau$ is the only scaling parameter. Both Cole-Cole plots show reflection symmetry about $\mathrm{Re}[|\mu - \mu_0|/\Delta\mu] = 0.5$, which is a consequence of the Cole-Cole distribution function.

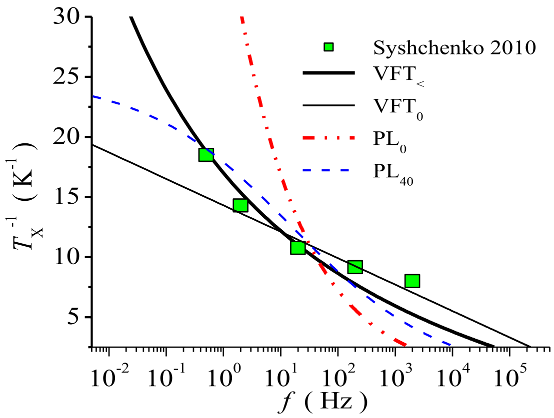

FIG. 3 (color online). Prediction for the inverse crossover temperature versus applied frequency. The green squares for 2, 20, and 200 Hz correspond to those marked in Fig. 1. The 0.5 and 2000 Hz data from [21] are also plotted for demonstration purposes. For power law with phase transition occurring at 40 mK, we used $\tau = \tau_0(|T_g|/(T - T_g))^p$ for $T > T_g$ and $\tau = \infty$ for $T \leq T_g$.

of the solid concomitant with a peak in dissipation. By studying the glass susceptibility due to the backaction on solid helium, we find that both the amplitude change and T dependence of the shear modulus are well captured by this model. An important consequence of the dynamic response analysis is the description of the dissipation or phase angle. In the proposed glass model, the peak height of the dissipation is independent of the applied frequency (in linear response) and linearly proportional to the Cole-Cole exponent α as well as the backaction strength g. Since g depends on the concentration of the TLS, we predict that increasing disorder will result in larger amplitude changes of the shear modulus. We hypothesize that the freezing out of fluctuating segments of dislocation lines are the relevant excitations contributing to the reported anomalies in solid ^{4}He. Additionally, we extracted a universal scaling behavior proportional to $\omega\tau(T)$. This corresponds to Cole-Cole plots collapsing onto a single curve over a wide range of frequencies. We would encourage future experiments to verify this prediction. Finally, we find that for a positive ideal glass temperature T_g the crossover temperature T_X converges toward T_g. This is another prediction that can serve as a clear experimental demonstration for the existence of a true phase transition in solid ^{4}He at low temperatures, if it exists. A detailed understanding of the microscopic nature of the glass and its excitations remains a pressing challenge. Future experimental characterizations may elucidate the puzzles found in the dynamics and thermodynamics of solid ^{4}He.

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