Two-level systems and mass deficit in quantum solids

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- A solid with a single quantum two-level system
- Adiabatic regime
- Finite-frequency response
- The case of incoherent tunnelling

Torsional oscillator experiments:

- Kim and Chan (2004, 2005, 2006)
- Rittner and Reppy (2006, 2007)
- Kondo, Takada, Shibayama, Shirahama (2007)
- Aoki, Graves and Kojima (2007)
- Clark, West and Chan (2007); Clark, Maynard and Chan (2008)
- Kim, Xia, West, Lin, Clark, Chan (2008)
- Hunt, Pratt, Gadagkar, Yamashita, Balatsky, Davis (2009)

Absence of mass flow:

- Greywall (1977)
- Bonfait, Godfrin, Castaing (1989)
- Day, Hermann, Beamish (2005); Day and Beamish (2006, 2007)

Observation of mass flow:

- Ray and Hallock (2008)

function for the ground state, as well as recent estimates (*8*, *9*) are in favor of there being vacancies at the 10^{-4} per site level, even in the pure crystal (SOM text). Repeated efforts by Clark *et al.* to grow perfect, pure, single crystals have always observed nonclassical rotational inertia (NCRI) on the 10^{-4} level relative to the classically expected value (*10*), and this level or higher has been confirmed by others (*11*–*13*). Simulations (*14*, *15*) performed using the path-integral Monte Carlo method have been claimed to prove the nonexistence of vacancies; however, among other difficulties the equivalent temperature in these simulations is well above the relevant temperature at which Bose condensation takes place. In any case, simulating 10^4 atoms well enough to find a single defect is beyond the capabilities of those methods. I used a background density ($|\Psi|^2$) relative to the solid for pure ^{4}He) of 2×10^{-4} to 3×10^{-4}, which fixes μ in terms of g.

I assumed m^* to be fairly light relative to that of a helium atom (m_{He}) and used an estimate that is often quoted,$1/3m_{\mathrm{He}}$ (other estimates are even smaller). This effective mass is such that the uncertainty energy that is necessary to localize the mass on a single site is on the order of 10 K. This is the same magnitude as estimates in (*10*) of the energy cost of a vacancy and suggests that those estimates may not have taken into account the kinetic energy that could be gained by delocalization. Regarding vacancies classically as strictly local configurations of the lattice is not reasonable.

m^* and the density of the boson field allow an estimate of the superfluid transition temperature from the Bose-Einstein equation

$$k_B T_c = \frac{2\pi h^2}{m^*}\left(\frac{N}{2.61V}\right)^{2/3} \quad (3)$$

where N/V is the vacancy density, k_B is Boltzmann's constant, and T_c is the transition temperature.

By entering into Eq. 3 a typical solid density, a vacancy concentration of 2×10^{-4} to 3×10^{-4} per site, and a mass of $1/3m_{\mathrm{He}}$, the resulting transition temperature is ~50 to 70 mK. This is very close to the transition temperature at which thermal hysteresis in the NCRI has been reported (*12*, *16*). I have discussed elsewhere (*17*) why reversible NCRI appears so far above T_c. One expects true superflow to be observable only below this T_c, if at all.

The parameter g, or equivalently the scattering length a, is not something one can accurately estimate. I next discuss here the consequences of assuming that g is reasonably small. This perhaps can be justified, again, from the fact that a light mass implies a somewhat extended lattice distortion. Here, I define a correlation length as

$$\xi = 1/\sqrt{8\pi n_0 a} \quad (4)$$

where $n_0 = |\Psi^2|$, which is the exponential decay length of a small perturbation in the vacancy field, according to Eq. 1. Given that $n_0 = 3 \times 10^{-4}$, even if a is a whole lattice constant, ξ is 10 lattice constants or 3 nm; it would be reasonable for ξ to be an order of magnitude larger. This is still not quite the scale at which the variation of surface-to-volume ratio in NCRI occurs (*18*), but almost.

What is of most interest is the effect of defects being attractive sites for vacancies. A dislocation core, for instance, is said to attract on the order of one vacancy per atomic length, calculated on the basis of localized high-energy vacancies (*19*). This amounts to a potential well in V that could be estimated as $VR^2 \approx 10$ to 15 K, where R is the well's radius. Balancing this against the repulsive interaction $g\Psi^4$, a dislocation core might be capable of attracting a cloud of $\propto 1/g$ vacancies with a radius on the order of ξ. Thus, the effect of dislocations can be somewhat magnified. Correspondingly, one would expect there to be similar diffuse densities of delocalized vacancies around grain boundaries and near surfaces. I would consider this to be one of the few possibile explanations for the degree to which crystal imperfection appears to enhance NCRI.

Why small concentrations of ^{3}He produce large effects remains an open question.

It seems possible to provide an accounting of most of the puzzling properties of low-temperature solid He by describing it as a Gross-Pitaevskii fluid of delocalized quantum vacancies. The idea that the superfluid is an intrinsic property of the pure crystal, which is locally enhanced by imperfections, seems to account for the low and reasonably invariant genuine superfluid transition and the large variations in the quantity of superflow, which otherwise appear to be irreconcilable.

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References

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Evidence for a Superglass State in Solid ^{4}He

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Although solid helium-4 (^{4}He) may be a supersolid, it also exhibits many phenomena unexpected in that context. We studied relaxation dynamics in the resonance frequency $f(T)$ and dissipation $D(T)$ of a torsional oscillator containing solid ^{4}He. With the appearance of the "supersolid" state, the relaxation times within $f(T)$ and $D(T)$ began to increase rapidly together. More importantly, the relaxation processes in both $D(T)$ and a component of $f(T)$ exhibited a complex synchronized ultraslow evolution toward equilibrium. Analysis using a generalized rotational susceptibility revealed that, while exhibiting these apparently glassy dynamics, the phenomena were quantitatively inconsistent with a simple excitation freeze-out transition because the variation in f was far too large. One possibility is that amorphous solid ^{4}He represents a new form of supersolid in which dynamical excitations within the solid control the superfluid phase stiffness.

A "classic" supersolid (*1*–*5*) is a bosonic crystal with an interpenetrating superfluid component. Solid ^{4}He has long been the focus of searches for this state (*6*). To demonstrate its existence unambiguously, macroscopic quantum phenomena (*7*) such as persistent mass currents, circulation quantization, quantized vortices, or the superfluid Josephson effect must be observed. None of these effects have been detected in solid ^{4}He.

There are, however, indications that this material could be a supersolid. This is because high-Q torsional oscillators (TOs) containing solid ^{4}He exhibit an increase in resonance frequency must always change in a quantitatively related manner. Such changes are measurable because

$$\frac{2(f_0 - f(T))}{f_0} = \frac{1}{I\omega_0^2}\Re[\chi_D^{-1}(T)]$$

$$D(T) - D_\infty = \frac{1}{I\omega_0^2}\Im[\chi_D^{-1}(T)] \qquad \text{(3A, 3B)}$$

within the Debye model with suitable approximations (*31*); $D_\infty \equiv \gamma/I\omega_0$. Moreover a well-defined characteristic temperature T^* for such a susceptibility occurs when $\omega_0\tau(T^*) = 1$; both the $f(T)$ slope and the dissipation $D(T)$ achieve their maxima at T^* (Fig. 1).

In Fig. 4A (left), we show a fit of Eq. 3B to the measured $D(T)$ as a red line, while Fig. 4A (right) shows the resulting prediction from Eq. 3A for $f(T)$ as the blue line. Comparison to the measured $f(T)$ (solid blue circles) shows that this Debye susceptibility is inconsistent with the relation between $D(T)$ and $f(T)$. Nevertheless, as the relaxation processes of $D(t,T)$ and $f(t,T)$ are synchronized (Fig. 3), there must be an intimate relation between $\Re[\chi^{-1}(t,T)]$ and $\Im[\chi^{-1}(t,T)]$. To study this relation, one should replot the data from Fig. 3, A and B, in the complex plane with axes defined by $\Im[\chi_D^{-1}]$ and $\Re[\chi_D^{-1}]$ [a Davidson-Cole (D-C) plot (*31*)]. This is a classic technique in which departures of the data from the Debye model appear as geometric features that can reveal characteristics of the underlying physical mechanism linking $\Re[\chi^{-1}(t,T)]$ and $\Im[\chi^{-1}(t,T)]$.

We therefore plot $\Delta D(T) = D(T) - D_\infty$ versus $\frac{2(f_0 - f(T))}{f_0}$ in Fig. 4B. It reveals that, instantaneously upon warming, the D-C plot is a symmetric elliptical curve, whereas after several thousand seconds, the response has evolved into the skewed D-C curve more familiar from studies of the dielectric glass transition (*34*). But the maximum frequency shift expected from the maximum observed dissipation within the Debye susceptibility (vertical dashed lines) is again far too small. Moreover, no temperature equilibration lag between the solid ^{4}He sample and the mixing chamber could generate the complex dynamics reported in Fig. 4 because, for any given frequency shift, a wide variety of different dissipations are observed [see supporting online material (SOM) text].

A simple superfluid transition is inconsistent with all these observations because there should be no synchronized dissipation peak associated with $f(T)$ (Figs. 1 and 4) and no ultraslow dynamics in $f(t,T)$ and $D(t,T)$ (Figs. 2 and 3). Indeed, these phenomena are more reminiscent of the characteristics of a glass transition (*34*). Nevertheless, a simple freeze-out of excitations described by a Debye susceptibility is also quantitatively inconsistent because the dissipation peak is far too small to explain the observed frequency shift (Fig. 4A). Thus, when considered in combination with implications of the blocked annulus experiments (*9*, *16*), our observations motivate a new hypothesis in which amorphous solid ^{4}He is a supersolid, but one whose superfluid phase-stiffness can be controlled by the freeze-out of an ensemble of excitations within the solid.

Within such a model, generation of excitations at higher temperatures would suppress superfluid phase stiffness. The complex relaxation dynamics (Figs. 3 and 4) would reveal the excitation freeze-out processes. Further, the anomalously large frequency shifts (Fig. 4) would occur predominantly because of superfluid phase stiffness appearing after excitation freezing. Such a model might also explain the diverse phenomenology of solid ^{4}He. For example, the ω dependence of T^* (*13*) would occur because T^* is the temperature for which $\tau(T^*)\omega = 1$. The shear modulus stiffening (*21*) would occur because of the freeze-out of motion of these excitations, and T^* would increase with ^{3}He concentration (*8*, *11*) because, with pinning, higher temperatures would be required to achieve the excitation rate $\tau(T^*)\omega_0 = 1$. Finally, sample-preparation effects (*10*, *12*) and different responses from different TO types would occur because the amorphousness allowing these excitations would depend on annealing and TO design.

Independent of these hypotheses, important new features of solid ^{4}He are revealed here. We find synchronized ultraslow relaxation dynamics of dissipation $D(T)$ and a component of frequency shift of $f(T)$ in TOs containing amorphous solid ^{4}He (Fig. 3). Such phenomena are reminiscent of the glassy freeze-out of an ensemble of excitations and inconsistent with a simple superfluid transition. Nevertheless, although the evolutions of $f(T)$ and $D(T)$ are linked dynamically, the situation is also inconsistent with the simple excitation freezing transition because there is an anomalously large frequency shift (Fig. 4). One possible explanation is that solid ^{4}He is not a supersolid and that the appropriate rotational susceptibility model for its transition will be identified eventually. But if superfluidity is the correct interpretation of blocked annulus experiments (*9*, *16*), then our results indicate that solid ^{4}He supports an exotic supersolid in which the glassy freeze-out at T^* of an unknown excitation within the amorphous solid controls the superfluid phase stiffness. Such a state could be designated a "superglass."

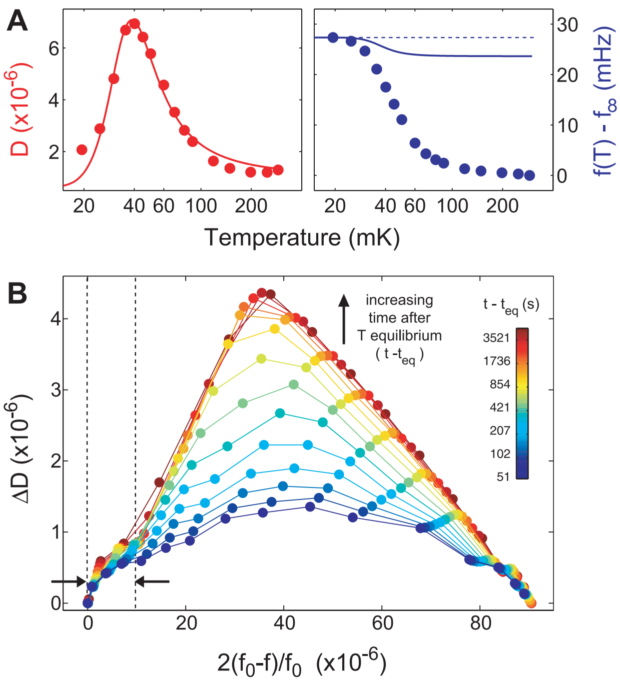

Fig. 4. (**A**) Comparison of equilibrated $D(T)$ and $f(T)$ data with simple Debye model of susceptibility in Eq. 1. The long-time equilibrated data $D(T)$ (left) and $f(T)$ (right) are plotted as filled circles. The dashed line indicates f_0. The solid curves represent the predicted relation between the susceptibilities for a simple glassy freeze-out transition. Although the dissipation $D(T)$ can be fit reasonably well by this model (*22*), the magnitude of the frequency shift that is then predicted is markedly smaller than observed. (**B**) Time-dependent D-C plot. This is a parametric plot of the data shown in Fig. 3, A and B, made by removing the explicit dependence on temperature, with axes defined by $\Im[\chi_D^{-1}]$ and $\Re[\chi_D^{-1}]$ (Eq. 3). The vertical dashed lines indicate the maximum value of $2(f_0 - f)/f_0$ that would be predicted by the Debye susceptibility (Eq. 1), given the peak height of $\Delta D = D - D_\infty$ (*31*).

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Classical theory of supersolids:

- Andreev and Lifshitz (1969); Chester (1970); Leggett (1970)

New theoretical developments:

- Prokofiev and Svistunov (2005); Pollet, Boninsegni, Kuklov, Prokof'ev, Svistunov, Troyer (2007) superfluidity of grain boundaries
- Andreev (2007) quantum two-level systems
- Anderson (2007) fluid of fluctuating quantum vortices
- Nussinov, Balatsky, Graf, Trugman (2007) "standard glass" response
- Yoo and Dorsey (2009) viscoelastic solid
- Anderson (2009) rarified Gross-Pitaevskii superfluid of vacancies

A SOLID WITH A SINGLE QUANTUM TWO-LEVEL SYSTEM

$$\widehat{H}_0 = \frac{1}{2M}\mathbf{P}^2 - \varepsilon\widehat{\sigma}_3 + J\widehat{\sigma}_1 \,,$$

2ε - energy difference, J - tunnelling amplitude.

The center of mass velocity:

$$\mathbf{V} \equiv \frac{d}{dt}\mathbf{R} = \frac{i}{\hbar}\left[H_0, \mathbf{R}\right] = \frac{1}{M}\mathbf{P}$$

is a classical variable commuting with $\widehat{H}_0$.

For $\widehat{H} = \widehat{H}_0 - \mathbf{F}\mathbf{R}$

$$\frac{d}{dt}\mathbf{V} = \frac{\mathbf{F}}{M}$$

The expression for the "coordinate" of the crystal lattice

$$\mathbf{X} = \mathbf{R} + \frac{m}{2M}\mathbf{a}\hat{\sigma}_3 \,.$$

follows from the definition of the center of mass position $\mathbf{R}$:

$$M\mathbf{R} = (M-m)\mathbf{X} + m\mathbf{r} = M\mathbf{X} + m(\mathbf{r} - \mathbf{X})\,,$$

with $\mathbf{u} \equiv \mathbf{r} - \mathbf{X}$ replaced by $-\frac{1}{2}\mathbf{a}\hat{\sigma}_3$.

If an external force $\mathbf{f}(t)$ is applied to the crystal lattice:

$$\hat{H} = \hat{H}_0 - \mathbf{X}\mathbf{f}(t) = -\mathbf{R}\mathbf{f}(t) - h_a(t)\hat{\sigma}_\alpha\,,$$

where $h_\alpha(t) = \left[-J,\ 0,\ \varepsilon + \frac{m}{2M}\mathbf{a}\mathbf{f}(t)\right]$, the crystal lattice velocity

$$\mathbf{v} \equiv \frac{d}{dt}\mathbf{X} = \frac{i}{\hbar}\left[\hat{H}_0, \mathbf{X}\right] = \frac{1}{M}\left(\mathbf{P} + \frac{mJ}{\hbar}\mathbf{a}\hat{\sigma}_2\right)$$

does not commute with $\hat{H}$.

$$\frac{d}{dt}\langle \mathbf{v} \rangle = \frac{\mathbf{f}}{M} + \frac{m}{M}\frac{J\mathbf{a}}{\hbar}\frac{d}{dt}\langle \sigma_2 \rangle \,,$$

ADIABATIC REGIME

$$\langle \sigma_\alpha \rangle \parallel h_\alpha \Rightarrow \langle \sigma_2 \rangle = 0 \Rightarrow M_{\mathrm{eff}} = M$$

Andreev (2007)

starts from $H_{\mathsf{TLS}} = -\varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1$
and applies the Galilean transformation to obtain

$$\hat{H} = \frac{M}{2}\mathbf{v}^2 - \frac{mJ}{\hbar}(\mathbf{av})\,\hat{\sigma}_2 - \varepsilon\hat{\sigma}_3 + J\hat{\sigma}_1 \quad \left(\equiv \frac{1}{2M}\mathbf{P}^2 + \hat{H}_{\mathsf{TLS}} + \mathsf{const}\right)$$

This Hamiltonian (with $\mathbf{v}$ treated as classical variable) gives for the TLS contribution to the total momentum $\mathbf{P} = M\mathbf{v} + \langle\mathbf{p}\rangle$:

$$\langle\mathbf{p}\rangle = -\left(\frac{mJ}{\hbar}\right)^2 \frac{\tanh[E(\mathbf{v})/T]}{E(\mathbf{v})}\,\mathbf{a}(\mathbf{av})$$

with

$$E(\mathbf{v}) \equiv \sqrt{\varepsilon^2 + J^2 + (mJ/\hbar)^2\,(\mathbf{av})^2}\,.$$

FINITE-FREQUENCY RESPONSE

$\mathbf{f}(t) = \mathbf{f}_0 \cos(\omega t)$. For vanishing (but non-zero) dissipation

$$\frac{d}{dt}\hat{\sigma}_\alpha = \frac{i}{\hbar}[H, \hat{\sigma}_\alpha] = -\frac{2}{\hbar}\epsilon_{\alpha\beta\gamma} h_\beta \hat{\sigma}_\gamma$$

solution of linearized equations for $\langle \hat{\sigma}_\alpha \rangle$ gives

$$\frac{d}{dt}\langle \hat{\sigma}_2 \rangle = -\frac{m}{M}\frac{\omega^2}{\omega^2 - \Omega^2}\frac{\mathbf{af}(t)}{\hbar}\langle \hat{\sigma}_1 \rangle^{(0)}$$

with $\Omega = 2E/\hbar$, $\langle \hat{\sigma}_1 \rangle^{(0)} = -(J/E)\tanh(E/T)$ and $E^2 = \varepsilon^2 + J^2$.

$$\frac{1}{M_{\text{eff}}(\omega)} = \frac{1}{M} + \frac{1}{3M^2}\sum_n \frac{\omega^2}{(\omega + i/\tau_n)^2 - \Omega_n^2}\,\lambda_n \, ,$$

where $$\lambda_n \equiv \frac{m_n^2 J_n^2 a_n^2}{\hbar^2 E_n}\tanh\frac{E_n}{T}$$

and τ_n - transverse relaxation time.

THE CASE OF INCOHERENT TUNNELLING

The classical analog of the equation for $d\mathbf{v}/dt$:

$$\frac{d}{dt}\mathbf{v} = \frac{1}{M}\left[\mathbf{f} - \frac{m}{M}\frac{d}{dt}\frac{d\mathbf{u}}{dt}\right],$$

Substitution of

$$\mathbf{u}(\omega) = -\frac{1}{-i\tau\omega + 1}\frac{m}{M}\frac{\mathbf{a}(\mathbf{af})}{4T\cosh^2(\epsilon/T)}$$

(Koshelev and Vinokur, 1991) gives

$$\frac{1}{M_{\mathrm{eff}}(\omega)} = \frac{1}{M} - \frac{\omega^2}{12TM^2}\sum_n \frac{1}{-i\tau_n\omega + 1}\tilde{\lambda}_n, \quad \tilde{\lambda}_n = \frac{m_n^2 a_n^2}{\cosh^2(\varepsilon_n/T)}$$

Torsional oscillator equation of motion

$$\left[K - i\gamma\omega - (I + I_{\mathrm{He}})\omega^2\right]\theta = 0$$

Nussinov, Balatsky, Graf, Trugman (2007);
Hunt, Pratt, Gadagkar, Yamashita, Balatsky, Davis (2009):

$$I_{\mathrm{He}}\omega^2 = I_0\omega^2 + \frac{g}{[1 - i\omega\tau(T)]}$$