PHYSICAL REVIEW B **75**, 094201 (2007)

Entropy of solid ^{4}He: The possible role of a dislocation-induced glass

A. V. Balatsky,[1,*] M. J. Graf,[1] Z. Nussinov,[2] and S. A. Trugman[1]
[1] *Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2] *Department of Physics, Washington University, St. Louis, Missouri 63160, USA*

(Received 18 December 2006; published 1 March 2007)

Solid ^{4}He is viewed as a nearly perfect Debye solid. Yet recent calorimetry indicates that its low-temperature specific heat has both cubic and linear contributions. These features appear in the same temperature range ($T \sim 200$ mK) where measurements of the torsional oscillator period suggest a supersolid transition. We analyze the specific heat to compare the measured with the estimated entropy for a proposed supersolid transition with 1% superfluid fraction. We find that the experimental entropy is substantially less than the calculated entropy. We suggest that the low-temperature linear term in the specific heat is due to a glassy state that develops at low temperatures and is caused by a distribution of tunneling systems in the crystal. It is proposed that small scale dislocation loops produce those tunneling systems. We argue that the reported mass decoupling is consistent with an increase in the oscillator frequency, as expected for a glasslike transition.

DOI: 10.1103/PhysRevB.75.094201 PACS number(s): 67.40.Kh, 05.30.Jp, 73.21.−b

I. INTRODUCTION

A supersolid is a novel state of matter that simultaneously displays both superfluidity and crystalline orders. ^{4}He is thought to be a most likely candidate for the supersolid state. Recent torsional oscillator experiments by Kim and Chan[1–4] generated renewed interest in this possibility. Pioneering work by Andreev and Lifshitz,[5] Reatto,[6] Chester and Reatto,[7] Chester,[8] Leggett,[9] and Anderson[10] laid the foundation for our knowledge about this enigmatic state. Recently, Anderson and co-workers revisited this problem,[11,12] and the latest developments, presented at the KITP workshop, are available online (see Ref. 13).

In addition to the work of the PSU group,[1–4] there is now an independent confirmation of the anomalous behavior of solid ^{4}He, as presented by Rittner and Reppy,[14] Shirahama,[15] and Kondo *et al.*[16] All groups use torsional oscillators similar to that used by the PSU group of Kim and Chan.[1–4] At the same time Rittner and Reppy[14] report a history dependence of the signal, when annealing the sample, to the extent of observing no mass decoupling in the torsional oscillator experiment. These torsional oscillator experiments clearly indicate anomalous mechanical properties of solid ^{4}He at low temperatures, similar to the acoustic wave and heat pulse experiments performed earlier by Goodkind and co-workers.[17–19] However, the relationship between the mechanical measurements and the suggested superfluidity is not direct. The most direct proof of superfluidity would be the observation of persistent current. In this regard, there is a recent experimental search for superflow by Day *et al.*,[20] which indicates no mass flow of any kind to very high accuracy. Thus, we conclude that the effect first observed by the PSU group is likely not an intrinsic property of solid ^{4}He because it depends critically on the ^{3}He concentration and shows annealing dependence.[14]

Alternatively, we suggest that many of the experimental facts seem to be consistent with a glasslike behavior of crystalline ^{4}He at temperatures $T \leqslant 200$ mK. For a lack of any definitive experiment, we assume that the glass in ^{4}He is formed due to dislocations. Recent data by Rittner and Reppy[14] indicated both strong history dependence of the signal on the sample quality and annealing of the effect, seen in torsional oscillator. Both observations are consistent with the defect and possible dislocation origin of the glassy state that we postulate. This picture is also consistent with the very recent report by Sasaki *et al.*[21] of anomalous flow in ^{4}He in the presence of significant amount of defects and grain boundaries. In this paper, we would like to decouple the discussion of the observed features in specific heat and torsional oscillator from the supersolidity. We will focus on two critical features reported to date by experiments. (1) The small entropy that is seen experimentally near the transition temperature, which is at least two orders of magnitude smaller than the expected entropy of a 1% supersolid fraction. (2) The observed linear specific heat in the *bosonic* crystal of ^{4}He, consistent with a glass, but not with a Debye crystal.

II. ENTROPY ANALYSIS

We focus on the specific heat measurements on ^{4}He by Clark and Chan,[22] which indicate a departure from the conventional T^3 specific heat behavior expected at low temperatures. Given the data, we searched for evidence of a thermodynamic phase transition to a supersolid phase, assuming a 1% concentration of condensate. The observed features in the specific heat occur in the same temperature range, where a change in the period of the torsional oscillator led to the speculation of a transition to a supersolid state (see Fig. 1). A low-temperature linear term in the specific heat can be discerned. We note that a linear term in the specific heat of ^{3}He and ^{4}He crystals was observed more than 40 years ago by Heltemes and Swenson[23] and Franck,[24] but not in later measurements by Edwards and Pandorf.[25] A theoretical explanation of the earlier results was given in terms of thermal vibrations of pinned dislocation segments based on the theory of Granato.[26]

We now perform a general thermodynamic analysis of the measured specific heat data by Clark and Chan (see Figs. 1 and 2). This is especially attractive for the analysis as its

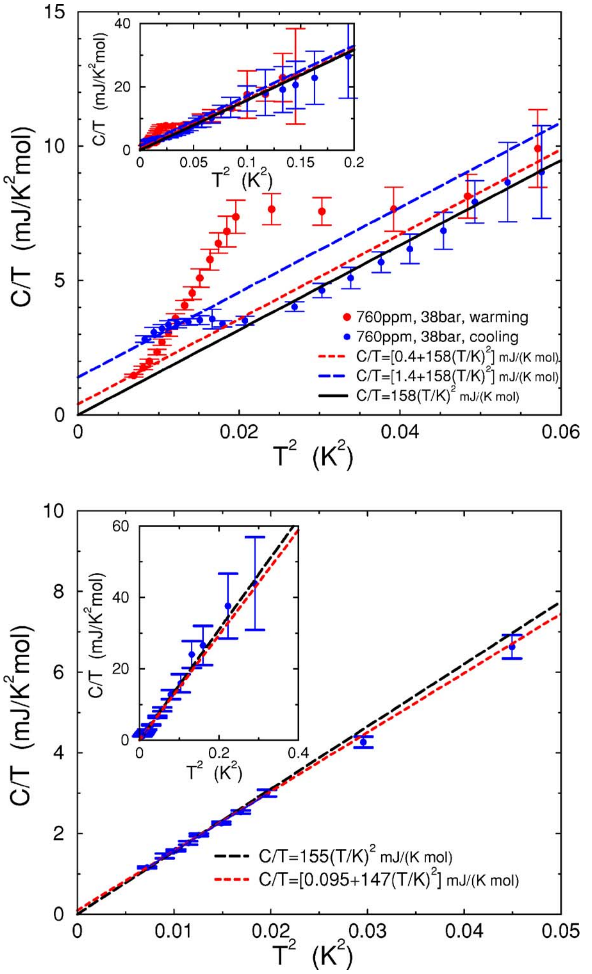

FIG. 1. (Color online) Specific heat over temperature of solid ^{4}He by Clark and Chan (Ref. 22). (Top) Sample with 760 ppm ^{3}He at 38 bars. (Bottom) Sample with 30 ppm ^{3}He at 39 bars. The dashed and solid lines are the respective low- and high-temperature fits, whose coefficients are presented in Table I. The insets show an enlarged temperature region.

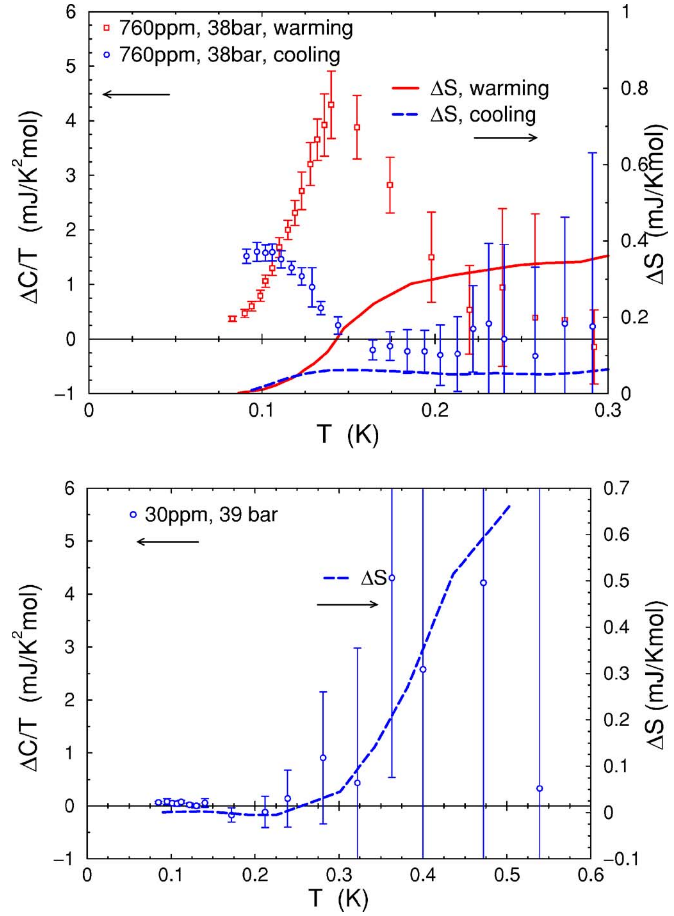

FIG. 2. (Color online) The $\Delta C/T$ differences and the corresponding entropies $\Delta S=\int_{T_{\min}}^{T} dT'\Delta C/T'$ at low T. (Top) Sample with 760 ppm of ^{3}He at 38 bars for cooling and warming runs; the high-T ($T>200$ mK) contribution of solid ^{4}He, $C=BT^3$, with $B_{760\ \mathrm{ppm}}=158$ mJ/(K^4 mol) was subtracted. (Bottom) Sample with 30 ppm of ^{3}He at 39 bars; the high-T contribution with $B_{30\ \mathrm{ppm}}=155$ mJ/(K^4 mol) was subtracted.

result will not depend sensitively on the exact functional form of the specific heat $C(T)$ at low temperatures, $T \leqslant 100$ mK. Most of the change in the entropy, $\int^T dT'[C(T')/T']$, which we find from the data, originates from the region of 100–400 mK. According to the torsion oscillator measurements, the supposed solid to supersolid transition occurs in the temperature range of 160–320 mK. The low-T entropy differences of the 760 and 30 ppm samples relative to solid ^{4}He are roughly $\Delta S(760\ \mathrm{ppm}) \sim 0.06-0.4$ mJ/(K mol) and $\Delta S(30\ \mathrm{ppm}) \sim 0.3$ mJ/(K mol), respectively, between 80 and 400 mK (see Fig. 2).[27]

Consistent with the experimental findings, we assume that roughly 1% of the bulk sample transforms into a superfluid; i.e., for simplicity, we assume that the condensate equals the superfluid fraction. Thus, 99% of the sample is unaffected and behaves like a perfect Debye crystal below ~500 mK ($T<\Theta_D/50$). Hence, the supersolid fraction would indicate an entropy of $S=46$ mJ/(K mol). We now compare the extracted entropy changes with what may be expected from a supersolid to solid transition (i) in a dilute gas of far apart and weakly interacting vacancies, as suggested by a 1% supersolid component, and (ii) in a strongly correlated dense gas: (i) The Bose-Einstein condensation (BEC) of an ideal gas gives values of entropy changes, which are an order of magnitude larger than those observed for the λ transition of liquid ^{4}He. We follow suit here and compare the ideal BEC with that of a possible supersolid transition, as is appropriate for a dilute gas of vacancies. Assuming a noninteracting Bose-Einstein gas in three dimensions with a parabolic band, the specific heat of the BEC is $C(T) = \frac{15}{4}[\zeta(5/2)/\zeta(3/2)]R(T/T_c)^{3/2}$ for $T \leqslant T_c$,[30] with $\zeta(z)$ the Riemann zeta function and R the gas constant. The total entropy per mole in the condensed state taken between $T=0$ and T_c is universally $S_{\mathrm{BEC}}(T_c)=\frac{5}{2}[\zeta(5/2)/\zeta(3/2)]R \approx 5R$. If we use a 1% molar concentration of supersolid fraction, we still get $S_{\mathrm{BEC}}(T_c) \approx 0.05R = 416$ mJ/(K mol) (per mole of ^{4}He). This is three orders of magnitude larger than the entropy changes seen in experiments (Fig. 2). These conclusions do not change if a short-range repulsive interatomic potential augments the kinetic energy in a dilute Bose gas. (ii) A lower bound can be found by scaling the measured S_λ

at the λ point of strongly correlated superfluid ^{4}He under pressure ($P_\lambda \sim 26$ bars), which is $S_\lambda = 4.6$ J/(K mol) at $T_\lambda = 1.8$ K.[31] As a generous lower bound on the entropy at a supersolid transition with strong correlations, we may correct for the fact that the claimed transition temperature is nine times smaller (~200 mK for 30 ppm sample) (Refs. 1 and 2) than T_λ. We scale down the expected 1% entropy by a factor of 9. Then, we may expect to find an entropy of $S_\lambda \sim 5$ mJ/(K mol). This very conservative estimate is about an order of magnitude bigger than the measured entropies. In ideal superfluids, a nearly T_c independent entropy $S(T_c)$ results, even for different low-T scaling regimes, e.g., phonons and protons. We emphasize that our linear scaling of $S(T_c)$ is a generous lower bound.[32]

More sensitive measurements of the specific heat using a redesigned experimental setup by Clark and Chan are consistent with the deviations from a T^3 behavior; however, the precise form at lowest temperatures, $T \leqslant 80$ mK, is not settled yet. Clearly, the estimated entropy associated with an actual superfluid transition is substantially larger than the measured $\Delta C/T$ integrated from ~80 to 500 mK for the case of λ transition or weakly interacting BEC. The observed deficit of entropy is very hard to reconcile with the 1% fraction of superfluid transition, which is suggested to account for the torsional oscillator experiments.

A null result of the signature of a λ transition, the observation of hysteresis on cooling and warming, and the dependence of the excess entropy on annealing point to a glasslike phenomenon of tunneling systems rather than that of a supersolid. Here, we note that recent analysis of the phase glass scenario is not directly applicable to the experiments and cannot explain the specific heat data.[33]

III. LINEAR SPECIFIC HEAT AND SENSITIVITY TO ^{3}He IMPURITIES

We propose that the linear specific heat term[22–24] is due to tunneling systems (TS) in the ^{4}He crystal. Specifically, we assume that dislocation loops are small enough to be present in large numbers ($>10^{10}$ cm^{-2}) to create the TS. We propose that fluctuations of nanoscale segments of dislocation loops form the TS. A small addition of ^{3}He atoms to the ^{4}He crystal facilitates the creation of dislocations; hence, the linear specific heat term should increase with ^{3}He concentration, similar to the effect of small amounts of hydrogen in the presence of dislocations in crystalline tantalum.[34]

The distribution of the characteristic energies of the TS is given by $P(E) = P_0 dE$, where we follow the standard discussion on the role of two-level systems in glasses.[28,29] It is commonly assumed that at low energy P_0 is constant. We assume that P_0 is only a function of the ^{3}He concentration n_3, and any contribution from intrinsic defects in ^{4}He is neglected. ^{3}He facilitates the creation of dislocations and hence P_0 should grow with the concentration of ^{3}He. We further assume that per mole of ^{4}He, $P_0(n_3) = cN_A n_3^\nu$, with positive exponent ν, coefficient c, and Avogadro's number N_A. A natural choice is $\nu = 1$, at least for low concentrations of ^{3}He. To keep the discussion general, we will not specify ν.

TABLE I. Summary of the linear and cubic coefficients of $C = AT + BT^3$ of solid ^{4}He with ^{3}He as solute, as well as its Debye temperatures Θ_D (Ref. 32). A is from the fits below ~120 mK, while B and Θ_D are from the fits between $200 < T \lesssim 500$ mK. Rough estimates of the uncertainties are in parentheses. For 30 ppm, A is close to zero as expected for this concentration.

^{3}He (ppm)	P (bar)	A [mJ/(K^2 mol)]	B [mJ/(K^4 mol)]	Θ_D (K)
30	39	0.09(4)	155(5)	23.2
760 (cooling)	38	1.4(2)	158(5)	23.1

The specific heat of a single tunneling system is $C_{\mathrm{TS}}(E,T) = k_B(E/k_BT)^2\{\exp(E/k_BT)/[1+\exp(E/k_BT)]^2\}$. The average over the distribution of the TS gives the total molar specific heat, which at low temperatures is $C_{\mathrm{TS}} = \int_0^\infty dEP(E)C_{\mathrm{TS}}(E,T) \approx (\pi^2/6)k_B^2P_0T$. The total specific heat of ^{4}He is the sum of the lattice (C_{lat}) and the TS (C_{TS}) contributions. For a perfect Debye crystal, the molar $C_{\mathrm{lat}} = (12\pi^4/5)R(T/\Theta_D)^3$ at low T. In solid ^{4}He, the Debye temperature $\Theta_D \approx 28$ K at $P \approx 40$ bars. Thus, at low temperatures ($T \lesssim \Theta_D/50 \approx 0.5$ K), the specific heat per mole of ^{4}He is $C = AT + BT^3$, with $A = (\pi^2/6)k_B^2P_0$ and $B = (12\pi^4/5)R/\Theta_D^3$. For $T < T^* = \sqrt{(\Theta_D^3k_B^2P_0)/(12\pi^2)} \sim n_3^{\nu/2}$, the linear term dominates over the lattice contribution. It follows that the TS model predicts that both the crossover temperature T^* and the linear coefficient A in the specific heat will be *very sensitive* with respect to ^{3}He concentration. For example, for $\nu = 1$, it leads to a square root dependence on n_3. This result also suggests that the effect of mass decoupling either vanishes or occurs at much lower temperatures in samples with vanishing ^{3}He concentration.

We revisited the data by Clark and Chan[22] (see Fig. 1), and assumed for our analysis linear and cubic terms in the specific heat. We find that the data are indeed consistent with a strong dependence on ^{3}He concentration. For example, the linear term depends on ^{3}He concentration, as shown in Table I. The A coefficient for ^{4}He reported by Heltemes and Swenson[23] and Franck[24] varied between $A \sim 2.5$ and 8.8 mJ/(mol K^2) at a similar pressure of $P \approx 40$ bars. They also reported a higher Debye temperature $\Theta_D \approx 28$ K as derived from Clark's data.[32] The A coefficients in the literature are somewhat bigger, but of the same order as ours, which are in the range of $A = 0.09 - 1.4$ mJ/(K^2 mol), depending on ^{3}He concentration and history.

The extracted low-temperature linear-term coefficient A scales roughly linearly with the ^{3}He concentration. Allowing for different functional forms for the specific heat for various systems of interacting bosons does not lead to a significant change in our results.

IV. THE "MISSING" MOMENT OF INERTIA AND SUSCEPTIBILITIES

The torsional oscillator experiments[1–4,14–16] measure the susceptibility—they do not directly monitor the moment of inertia of the supersolid, as discussed in depth in Ref. 35. As in any time translationally invariant system, the Fourier amplitude of the angular response of the torsion oscillator is $\theta(\omega)=\chi(\omega)\tau_{\text{ext}}(\omega)$, with $\chi=\chi_1+i\chi_2$ the angular susceptibility and τ_{ext} the external torque. For the simple torsional oscillator, $\chi^{-1}(\omega,T)=[\alpha-i\omega\gamma_{\text{osc}}-I_{\text{osc}}\omega^2+g(\omega,T)]$. Here, I_{osc} is the moment of inertia of the torsional oscillator, α is its restoring constant, γ_{osc} is the dissipative coefficient of the oscillator, and $g(\omega,T)$ arises from the back action of solid ^{4}He. For an ideal normal solid with moment of inertia I_{ns}, which rotates with the oscillator, the back action is $g_{ss}=-I_{ns}(T)\omega^2$. However, we do not need to impose this form. Current experiments measure the oscillator period $2\pi/\omega_0$, with ω_0 the real part of the solution of $\chi^{-1}(\omega,T)=0$ at fixed T. For example, a decrease in an effective dissipative component ($g_{\text{glass}}=-i\gamma_{\text{glass}}\omega$) in $g(\omega,T)$ as the temperature is lowered (wherein a liquid component transforms into a solid glass) will also lead to a shorter rotation period. The decrease in the rotation period only implies a crossover in χ (and a constraint on g). As the real and imaginary parts of χ are related by the Kramers-Kronig relations, an enhanced decrease in $\chi_1(\omega,T)$ often appears with a pronounced peak in χ_2 (as in Ref. 14). A nonvanishing χ_2 at finite frequency mandates dissipation. The simple forms g_{ss} and g_{glass} lead to different predictions for the dissipation, given the oscillator period which can be tested in theory.[36]

The microscopic deformation of the glass to the applied torsion might, similar to suggestions[37] concerning torsional oscillator results on granular media,[38,39] take the form of elastic, plastic, and fracture processes (including those of internal avalanches). A gradual change can proceed through dislocation glide in the slip plane.[40] All of these possibilities need to be addressed experimentally before we can have a definitive microscopic picture of the possible glass state. Neutron-scattering experiments may measure dislocation densities.[41] To test our theory for a dislocated solid ^{4}He, such experiments should be done within the pressure cell.[42]

One way to differentiate between glassy effects and a true thermodynamic ρ_s is to measure the frequency dependence of ρ_s. For the dislocation TS that we propose, $\rho_s(T,\omega)$ will be a nontrivial function of frequency, which vanishes as a power of frequency $\rho_S(T\rightarrow 0,\omega)\sim\omega^{\alpha}$. On the other hand, for a true supersolid phase there is a finite limit $\rho_S(T\rightarrow 0,\omega)\sim\rho_s(0)$.

V. HEAT PULSE EXPERIMENT

A heat pulse experiment may test the existence and relevance of the TS for the thermal properties.[43] In the absence of TS, the heat pulse would trigger quick equilibration of the energy with the phonon bath. Hence, on a very short time scale the temperature of a sample will reach a steady-state value. By contrast, for a crystal with TS, the heat deposited in the crystal will first be absorbed by the phonon bath and will later cause the repopulation of the TS as a result of the energy transfer from phonon bath to TS.

VI. CONCLUSIONS

We find that the measured entropy excess is several orders of magnitude smaller than the entropy expected from the BEC or λ transition of 1% superfluid fraction. While the absence of the entropy released at claimed supersolid transition is puzzling, it is consistent with a dislocation induced glassy state in ^{4}He crystals. This hypothesis predicts that the linear term in the specific heat increases with ^{3}He concentration. Heat pulse, heat transport, and ultrasound measurements would be helpful in identifying tunneling systems if they indeed exist in crystalline ^{4}He. The increase in the oscillator frequency at low temperatures is consistent with this interpretation.

Recently, we learned of melting curve data by Todoschchenko *et al.*,[44] which shows deviations from the Debye behavior and further supports our claim of thermodynamic measures inconsistent with a supersolid transition.

ACKNOWLEDGMENTS

The authors acknowledge discussions with E. Abrahams, P. W. Anderson, D. Arovas, C. D. Batista, B. Brinkman, M. Chan, D. Ceperly, A. Dorsey, D. Huse, J. Goodkind, A. J. Leggett, J. Reppy, and G. Zimanyi. This work was supported by U.S. DOE. A.V.B. is grateful to the KITP for its hospitality during the Supersolid Mini-Program.

---

*Electronic address: avb@lanl.gov

[1] E. Kim and M. H. W. Chan, Nature (London) **427**, 225 (2004).

[2] E. Kim and M. H. W. Chan, Science **305**, 1941 (2005).

[3] E. Kim and M. H. W. Chan, J. Low Temp. Phys. **138**, 859 (2005).

[4] E. Kim and M. H. W. Chan, Phys. Rev. Lett. **97**, 115302 (2006).

[5] A. F. Andreev and I. M. Lifshitz, Sov. Phys. JETP **29**, 1107 (1969).

[6] L. Reatto, Phys. Rev. **183**, 334 (1969).

[7] G. V. Chester and L. Reatto, Phys. Rev. **155**, 88 (1967).

[8] G. V. Chester, Phys. Rev. A **2**, 256 (1970).

[9] A. J. Leggett, Phys. Rev. Lett. **25**, 1543 (1970).

[10] P. W. Anderson, *Basic Notions of Condensed Matter Physics* (Benjamin, Menlo Park, CA, 1984), Chap. 4, p. 143.

[11] P. W. Anderson, cond-mat/0504731 (unpublished).

[12] P. W. Anderson, W. F. Brinkman, and D. A. Huse, Science **310**, 1164 (2005).

[13] KITP workshop presentations are available online at http://online.itp.ucsb.edu/online/smatter_m06

[14] Ann Sophie C. Rittner and J. D. Reppy, Phys. Rev. Lett. **97**, 165301 (2006).

[15] K. Shirahama, http://online.itp.ucsb.edu/online/smatter_m06/shirahama

[16] M. Kondo, S. Takada, Y. Shibayama, and K. Shirahama, cond-mat/0607032 (to be published).

[17] J. M. Goodkind, Phys. Rev. Lett. **89**, 095301 (2002).

[18] G. Lengua and J. M. Goodkind, J. Low Temp. Phys. **79**, 251 (1990).

[19] P.-C. Ho, I. P. Bindloss, and J. M. Goodkind, J. Low Temp. Phys.

109, 409 (1997).

[20] J. Day, T. Herman, and J. Beamish, Phys. Rev. Lett. **95**, 035301 (2005).

[21] S. Sasaki, R. Ishiguro, F. Caupin, H. J. Maris, and S. Balibar, Science **313**, 1098 (2006).

[22] A. C. Clark and M. H. W. Chan, J. Low Temp. Phys. **138**, 853 (2005).

[23] E. C. Heltemes and C. A. Swenson, Phys. Rev. **128**, 1512 (1962).

[24] J. P. Franck, Phys. Lett. **11**, 208 (1964).

[25] D. O. Edwards and R. C. Pandorf, Phys. Rev. **140**, A816 (1965).

[26] A. Granato, Phys. Rev. **111**, 740 (1958).

[27] Instead of integrating $\Delta C/T$, one could alternatively directly compare the function $\Delta C(T)/T$ to what is expected for the BEC. In Fig. 1 (top, cooling), the low-temperature bump has nearly the expected form for the BEC, but is approximately 500 times too small.

[28] P. W. Anderson, B. I. Halperin, and C. M. Varma, Philos. Mag. **25**, 1 (1972).

[29] W. A. Phillips, J. Low Temp. Phys. **11**, 757 (1972).

[30] L. Pitaevskii and S. Stringari, *Bose Einstein Condensation* (Clarendon, Oxford, 2003).

[31] G. Ahlers, Phys. Rev. A **8**, 530 (1973).

[32] In a private communication, A. C. Clark informed us that the heat capacity in Ref. 22 was overestimated by a factor 1.8. Including this factor leads to good agreement with literature values for Θ_D and supports even more our arguments for lack of entropy. However, it was not included in our analysis. Clark and Chan also reported hysteresis in C around T_c, which was assigned to phase separation of ^{3}He$-$^{4}He mixture in the 760 ppm sample. Regardless of a phase separation, our analysis indicates an entropy inconsistent with a 1% supersolid component.

[33] M. Boninsegni, N. Prokofev, and B. Svistunov, Phys. Rev. Lett. **96**, 105301 (2006).

[34] R. König, F. Mrowka, I. Usherov-Marshak, P. Esquinazi, and W. Wasserbäch, Physica B **316-317**, 539 (2002).

[35] Z. Nussinov, A. V. Balatsky, M. J. Graf, and S. A. Trugman, cond-mat/0610743 (to be published).

[36] For a general damped oscillator the period is $4\pi I/\sqrt{4\alpha I-\gamma^2}$ and the damping rate is $\gamma/(2I)$. For a simple glass embodied by g_{glass}, the dissipation $\gamma=\gamma_{\text{glass}}+\gamma_{\text{osc}}$ varies with temperature, while $I=I_{\text{osc}}+I_{ns}$ is fixed. Thus, a measured value of the period can be used to compute $\gamma(T)$ and then predict the dissipation and compare it to the experiment. Alternatively, for a supersolid, $g=g_{ss}$, then γ is fixed, and $I(T)$ varies. The predictions of both theories can be compared with experiments: (i) a glass transition with a simplified $g=g_{\text{glass}}$ and (ii) a purported supersolid transition with $g=g_{ss}$. Regretfully, so far, the experimental results of Refs. 1, 2, and 14 for the dissipation rates disagree.

[37] G. D'Anna, P. Mayor, G. Gremaud, A. Barrat, and V. Loreto, Europhys. Lett. **61**, 60 (2003).

[38] G. D'Anna and G. Gremaud, Nature (London) **413**, 407 (2001).

[39] G. D'Anna, P. Mayor, A. Barrat, V. Loreto, and F. Nori, Nature (London) **424**, 909 (2003).

[40] Y. Hiki and F. Tsuruoka, Phys. Rev. B **27**, 696 (1983).

[41] B. J. Heuser, J. Appl. Crystallogr. **27**, 1020 (1994).

[42] W. Bao, C. Broholm, and J. F. Trevino, Rev. Sci. Instrum. **66**, 1260 (1995).

[43] A. Nittke, S. Sahling, and P. Esquinazi, in *Tunneling Systems in Amorphous and Crystalline Solids*, edited by P. Esquinazi (Springer, Berlin, 1998), p. 9.

[44] I. A. Todoshchenko, H. Alles, H. J. Junes, A. Ya. Parshin, and V. Tsepelin, Phys. Rev. Lett. **97**, 165302 (2006).