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Supersolidity and Superfluidity of Grain Boundaries

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Abstract We have looked for dc mass transport through solid ^{4}He in a simple experiment with two communicating vessels filled with solid ^{4}He in equilibrium with liquid ^{4}He. Through good quality crystals, we have observed no mass transport, in contradiction with the hypothesis of a Bose–Einstein condensation of vacancies. Through crystals containing grain boundaries, we have found superfluid flow along these grain boundaries. We discuss these results in the context of other experiments on supersolidity.

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We present new evidence for the "supersolid" behavior which was recently discovered by Kim and Chan (KC) and confirmed by three other groups [1–5]. KC's experiment has triggered a considerable interest because it could be interpreted in terms of a Bose–Einstein condensation (BEC) of vacancies present in the crystals. If confirmed, it would be a remarkable coexistence of order in real space (crystalline order) and in momentum space (BEC leading to superfluidity). However this interpretation is controversial [6–12]. Furthermore, Rittner and Reppy [3] have found that annealing crystals destroys supersolidity. This is why we looked for effects of disorder in a different experiment. We have found mass transport along grain boundaries, which

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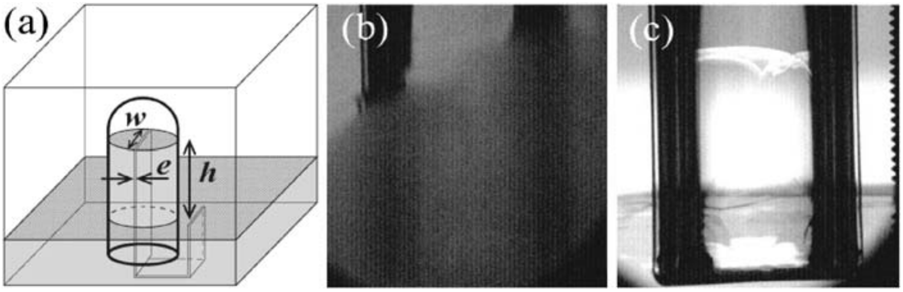

Fig. 1 **a** The cell contains a 1 cm diameter test tube and is partially filled with solid helium in equilibrium with liquid helium above. The solid inside the tube is higher by h than outside. Schematic grain boundaries (width w and thickness e) connecting the inner and the outer liquid are also shown. **b** A crystal grown from normal liquid ^{4}He at 1.8 K. **c** A crystal (# 3) with cusps at the liquid–solid interface indicating the existence of grain boundaries

are superfluid as proposed by Burovski et al. [6]. This mechanism is very different from the BEC of vacancies invoked by many authors [7, 13–15].

The principle of our experiment is simple: we have placed a glass tube (inner diameter $D = 1$ cm) in the cell of our optical cryostat which we filled with solid ^{4}He in equilibrium with superfluid ^{4}He above. The top of the tube is closed and we could bring the liquid–solid interface higher inside the tube than outside (Fig. 1a). Then we watched the possible relaxation of the height difference between the inside and the outside of the tube. Since the liquid density ρ_L is smaller than the solid density ρ_C, any level change inside the tube required mass transport through the solid. According to KC, a superfluid density ($\rho_s = 0.01\ \rho_C$) flows at a critical velocity $v_c = 10$ μm/s below 50 mK. These numbers correspond to natural ^{4}He containing about 0.1 ppm of ^{3}He, which is what we also used. We thus expected the height difference to relax at a velocity $V = \rho_s v_c/(\rho_C - \rho_L) = 1$ μm/s. As explained below, this is *not* what we have found.

In order to grow the solid inside the tube, we had to apply a substantial pressure difference from the outside. For this we had the inside of the tube at 1.3 K (consequently at the melting pressure $P_m(1.3\ \text{K}) = 25.7$ bar) and the outside at 1.4 K (and at $P_m(1.4\ \text{K}) = 26.0$ bar) for about 10 s. To do it, we used a heater or temporarily increased the flow rate through the fill line. After such a treatment, several cusps often appeared at the liquid–solid interface (Fig. 1c), meaning that grain boundaries (GB) had formed inside the crystal. These cusps result from the mechanical equilibrium between the GB surface tension and the liquid–solid interfacial tension. Once solid helium had entered the tube, we could cool down to 50 mK, lower the outside level 1 cm below the inside level by opening a valve on the fill line and watch the relaxation with a video camera.

We studied 13 crystals. For most of them, no cusps could be seen inside the tube, and we found no relaxation of the level within 50 μm over 4 hours, meaning that the interface velocity V was smaller than 3.5×10^{-3} μm/s. This observation rules out simple interpretations of KC's experiment in terms of 1% of the crystal mass being a 3D superfluid of vacancies with a critical velocity of 10 μm/s as mentioned above. It also puts a constraint on models involving a superfluid layer near the glass wall as proposed by Dash and Wettlaufer [10] and by Khairallah and Ceperley [11]. If this layer had a thickness e, a superfluid density ρ_s and a critical velocity v_c^W, we would find

$$v_c^W = \frac{D}{4e}\frac{\rho_C - \rho_L}{\rho_s} V < 0.28\frac{a}{e}\frac{\rho_C}{\rho_s}\ \text{cm/s}. \tag{1}$$

Dash explained KC's results with a value of e that is 4 to 8 times $a = 3$ Å, the thickness of one atomic layer. This is hardly compatible with (1) because such a thick layer should be similar to the films studied by Telschow [16] who measured superflow at 2 m/s. Khairallah proposed that $e = a$ and $\rho_s/\rho_C = 0.04$; this is possible if $v_c^W < 7$ cm/s, but then the mass flux looks too small to explain KC's experiment.

For three crystals (# 1, 2 and 3), we could see cusps at the liquid–solid interface inside the tube and the inner level relaxed (Fig. 2). Since crystals without cusps never relaxed, we attribute mass transport in these three crystals to GBs. Crystal 1 had only one visible cusp and the height $h(t)$ inside the tube relaxed by 0.8 mm. We checked that the relaxation stopped when the cusp disappeared on the left side. This means that there was only one GB at this place. The level moved at a velocity $V = 0.6$ μm/s, which was nearly constant; this is characteristic of superfluid flow at a critical velocity. Assuming that superfluid flow took place along one GB with a thickness e, a width w and a superfluid density ρ_s the critical velocity v_c^{GB} inside it was

$$v_c^{\text{GB}} = \frac{\pi D^2}{4ew}\frac{\rho_C - \rho_L}{\rho_s} V = 1.5\frac{aD}{ew}\frac{\rho_C}{\rho_s}\ \text{m/s}. \tag{2}$$

Since D/w and e/a have to be slightly larger than, but of order 1, (2) is compatible with Telschow's measurements of critical velocities in free liquid films (2 m/s for thicknesses of a few atomic layers). If GBs were not superfluid, the relaxation would be exponential and extremely slow. Note also that, if the top of the tube was open, the interface would relax with a very short time constant (0.2 μs) [17]. Thus, the observed relaxation is not limited by the melting dynamics.

Crystal 2 showed many cusps, and the height $h(t)$ relaxed about ten times faster than for crystal 1. It relaxed to zero and stayed there, showing that there was no temperature difference between the inside and the outside of the tube.

When we injected more helium into the cell, we increased the level outside but not inside where it was blocked by facets. In 1989, Bonfait et al. [18] tried an experiment similar to ours where the inner space of a cylindrical capacitor never filled in, and it was probably due to the same phenomenon, i.e. the faceting blocked the growth. This is why we studied supersolidity by looking at the melting of crystals, which eliminates facets from the solid surface.

Crystal 3 is the only one we studied at high temperature (1.13 K). It had rather inhomogeneous disorder and the relaxation rate changed several times. It reached equilibrium ($h = 0$) at a constant velocity similar to that of crystal 1 (Fig. 2c). We thus understand that GBs are superfluid up to 1.13 K at least and the superfluid transition temperature, T_c^{GB}, is higher than 1.13 K, which probably means that they are at least one atomic layer thick at the melting pressure P_m.

We also observed that GBs move transversely so that some cusps disappear in a time of order one hour. This must be due to local crystallization and melting triggered by the evolution of stress gradients. Some of the GBs must be pinned to the walls;

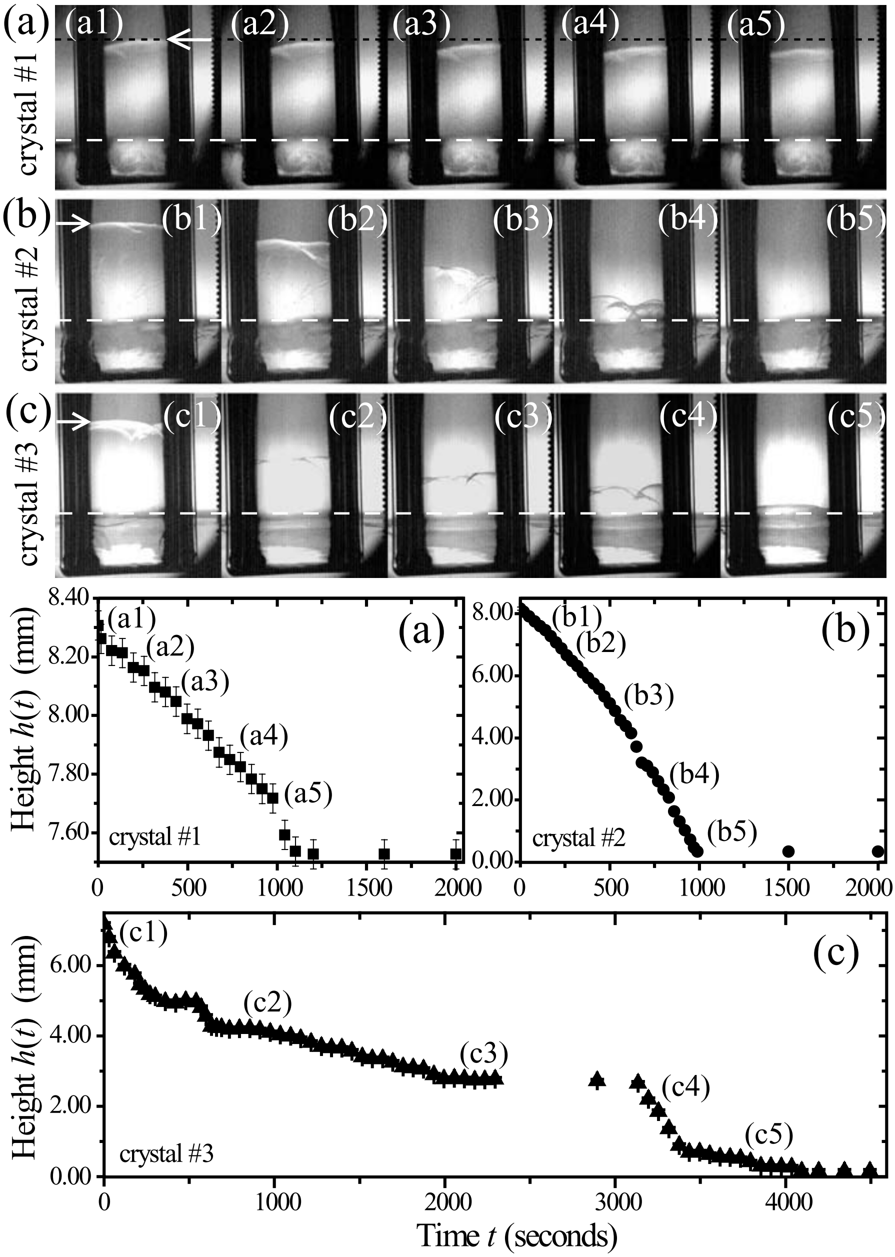

Fig. 2 Pictures and graphs of the relaxation of crystals 1 at 50 mK (**a**), 2 at 50 mK (**b**), and 3 at 1.13 K (**c**). Labels on graphs refer to pictures. The *white arrows* in *a1*, *b1* and *c1* indicate the position where the level was measured

some others move and vanish on the side walls or at the liquid–solid interface. As a consequence, the network of GBs can stop connecting the two sides of the crystal. The transverse motion explains the sudden stop in the relaxation of crystal 1, also why most of our crystals did not relax: within the few hours necessary to reach 50 mK, many GBs disappeared.

We could also make very bad quality crystals by growing them above 1.8 K. The liquid being normal, temperature gradients led to dendritic growth; snow flakes accumulating at the bottom of the cell formed a highly disordered solid which strongly scattered light (Fig. 1b). Unfortunately we could not fill the cell completely with such very bad quality crystals because the fill line always blocked before all the helium was solidified and we could not apply more than 34 bar in the cell. Since some liquid remained, the solid recrystallized as a good quality crystal when cooling through the bcc–hcp transition at 1.46 K. We could not study supersolidity in these crystals.

In summary, we have shown that grain boundaries are superfluid so that ^{4}He crystals with enough disorder are "supersolid". But they are not "supercrystalline" because mass superflow does not occur through the crystal itself. It is tempting to interpret KC's results in terms of GBs. Their "blocked capillary" method of growth should lead to many GBs, especially if the solid grows from a normal liquid. KC's 1% superfluid density would require a very high density of GBs (one every 100e). Assuming this, one could imagine that, in their experiment, there is a superfluid flow along connected GBs, so that the corresponding mass decouples from the torsional oscillator. KC observed that $\rho_s(P)$ increases up to 55 bar before decreasing at higher pressure; the increase could be due to an increasing GB density and the decrease to superfluidity vanishing near 200 bar [19]. However, further measurements are needed before KC's results can really be connected to ours, especially the superfluid transition temperature of grain boundaries as a function of pressure: GBs could be thicker at P_m than at high P where their superfluid density and their critical velocity might be much smaller if their thickness e is less than or comparable to one atomic layer. The adsorption of ^{3}He impurities on GBs has also to be measured.

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References

1. E. Kim, M.H.W. Chan, Nature **427**, 225 (2004)
2. E. Kim, M.H.W. Chan, Science **305**, 1941 (2004)
3. A.S.C. Rittner, J.D. Reppy, Phys. Rev. Lett. **97**, 165301 (2006)
4. K. Shirahama, M. Kondo, S. Takada, Y. Shibayama, Communication G41 7 at APS Conference, Baltimore, March 2006
5. M. Kubota et al., Science **311**, 1693 (2006), mentioned by A. Cho
6. E. Burovski, E. Kozik, A. Kuklov, N. Prokof'ev, B. Svistunov, Phys. Rev. Lett. **94**, 165301 (2005)
7. D.E. Galli, L. Reatto, Phys. Rev. Lett. **96**, 165301 (2006)
8. B.K. Clark, D.M. Ceperley, Phys. Rev. Lett. **96**, 105302 (2006)
9. M. Boninsegni, N. Prokof'ev, B. Svistunov, Phys. Rev. Lett. **96**, 105301 (2006)
10. G. Dash, J.S. Wettlaufer, Phys. Rev. Lett. **94**, 235301 (2005)
11. S.A. Khairallah, D.M. Ceperley, Phys. Rev. Lett. **95**, 185301 (2005)
12. P.W. Anderson, W.F. Brinkman, D.A. Huse, Science **310**, 1164 (2005)

13. A.F. Andreev, I.M. Lifshitz, Sov. Phys. JETP **29**, 1107 (1969)
14. G.V. Chester, Phys. Rev. A **2**, 256 (1970)
15. A.J. Leggett, Phys. Rev. Lett. **25**, 1543 (1970)
16. K. Telschow, I. Rudnick, T.G. Wang, Phys. Rev. Lett. **32**, 1292 (1974)
17. S. Balibar, H. Alles, A.Y. Parshin, Rev. Mod. Phys. **77**, 317 (2005)
18. G. Bonfait, H. Godfrin, B. Castaing, J. Phys. (Paris) **50**, 1997 (1989)
19. R. Ishiguro, F. Caupin, S. Balibar, Europhys. Lett. **75**, 91 (2006), and references therein