Local Stress and Superfluid Properties of Solid ^{4}He

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We provide a semiquantitative tool, derived from first-principles simulations, for answering the question of whether certain types of defects in solid ^{4}He support mass superflow. Although ideal crystals of ^{4}He are not supersolid, the gap for vacancy creation closes when applying a moderate stress. While a homogeneous system becomes unstable at this point, the stressed core of crystalline defects (dislocations and grain boundaries) can turn superfluid.

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The first microscopic mechanism of supersolidity was proposed by Andreev and Lifshitz [1] and Chester [2] arguing that a dilute gas of vacancies could lower the energy of an ideal helium crystal by quantum mechanically delocalizing the vacancies. At low temperature the vacancies undergo Bose-Einstein condensation and give rise to a supersolid—indeed, vacancies are required for supersolidity [3]. Although the detection of frictionless mass flow was anticipated to be straightforward, all experimental attempts failed for over 30 years [4] until Kim and Chan reported the observation of an unexpected drop of the torsional oscillator period at low temperatures [5]. Since the period is proportional to the square root of the moment of inertia, the origin of the period shift was attributed to the supersolid decoupling of the ^{4}He solid mass from the rotating walls. The basic effect is now confirmed by several groups [6–9], but the underlying physics remains unclear, both theoretically and experimentally [10].

First-principles calculations [11,12] rule out the possibility of supersolidity in ideal equilibrium crystals of ^{4}He. On the experimental side, no steady flow of mass through the solid phase was detected in several setups [13,14]. It is now believed that the superflow detected at the melting curve in Ref. [15] was due to liquid channels forming on the cell walls along the lines of contact with grain boundaries [10].

An interesting development is the observation of the liquid pressure equilibration by flow through a solid in the "UMass sandwich" experiment reported by Ray and Hallock [16] away from the melting curve. The dependence of flow on pressure difference was characteristic of critical superflow and inconsistent with the flow of a viscous liquid. We believe that an alternative interpretation of the experiment by mass flow through liquid channels is inconsistent with the absence of flow above $T > 0.6$ K, nor can a few liquid channels account for the observed mass flux value. Ray and Hallock argue that superfluidity along dislocations or grain boundaries can quantitatively account for their observations. In this Letter, we present semiquantitative arguments, based on first-principle simulations, which relates superfluidity of defects to large local strain.

In the past, several authors have speculated about the possibility of the stress-induced supersolidity [17–19], especially under hydrostatic decompression since quantum effects and vacancy delocalization are expected to increase at lower densities. The phenomenological models missed the attraction between vacancies which destabilizes a dilute homogeneous gas of vacancies. Central questions were left unanswered: What type and strength of stress is needed to close the insulating gap in ^{4}He and is this stress realistic?

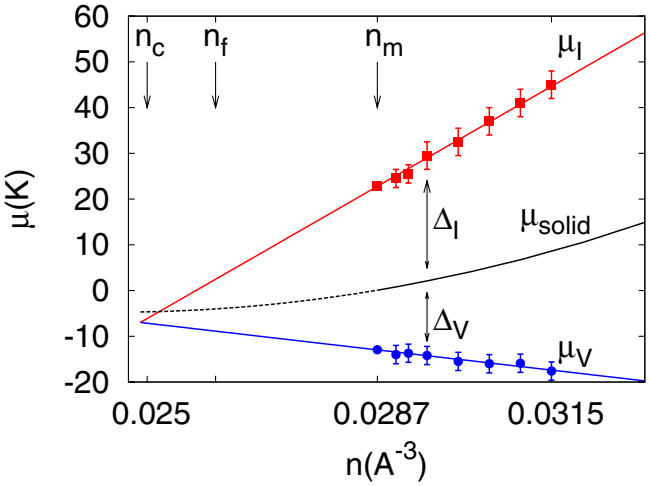

FIG. 1 (color online). Extrapolation of vacancy (Δ_V) and interstitial (Δ_I) gaps shows that the density corresponding to closing the gap lies in the liquid phase. The data points as a function of density for the ideal hcp solid are taken from Ref. [21]. By μ_V (μ_I) we denote the threshold chemical potentials for doping the system with vacancies (interstitials).

Our approach to this inherently strongly correlated problem is numerical. Feynman's path-integral formulation of quantum mechanics allows an exact mapping between the ^{4}He system and a system of world lines in four-dimensional space. The fourth dimension represents evolution in imaginary time and its extent is $\hbar/k_BT$ where $\hbar$ is Planck's constant, k_B is the Boltzmann constant, and T is temperature. This system of world lines is simulated efficiently by the worm algorithm [20]. Energy gaps for vacancy and interstitial excitations are readily obtained from the exponential decay in imaginary time of the one-body Green's function of the system [21].

Hydrostatic decompression.—After initial attempts to detect the supersolid state in ^{4}He failed, it was suggested that a metastable supersolid can form in crystals decompressed below their melting density of $n_m = 0.0287$ Å^{-3} (the freezing density is $n_f = 0.0261$ Å^{-3}). The idea turned out to be impossible to realize experimentally, and now we understand why it was implausible in the first place: in Fig. 1 we show the density dependence of the vacancy and interstitial gaps. Data are extrapolated to lower densities $n < n_m$ using the near perfect linear density dependence. The metastable hcp crystals remain insulating all the way to liquid densities and even beyond. It is unlikely that the solid structure will survive for long at liquid densities, and neither will the possible supersolid phase at density $n_c \approx 0.025$ Å^{-3}. The hydrostatic strain required to reach this density is about $(n_m/n_c - 1)^{1/3} \approx 13.5\%$. Assuming solid compressibility at melting [22] the required underpressure is close to -25 bar.

The (meta)stability of the supersolid phase depends on the sign of the effective interactions between vacancies. An attractive vacancy gas will collapse to the lower density liquid phase. To determine the sign and strength of the vacancy-vacancy interaction one has to know their mass m^* and their pair correlation function $\nu(r)$. In the ideal gas $\nu(r)$ is enhanced at short distances by a factor of 2 relative to the large distance limit. Correspondingly, for repulsive (attractive) interactions $\nu(r)$ is suppressed (enhanced) relative to the ideal gas behavior. If attractive interactions are so strong that vacancies actually form a bound state, then $\nu(r)$ is enhanced exponentially, and the spatial decay of the correlation function can be used to determine the binding energy from $\nu(r) \propto \exp(-2\sqrt{m^* E_b} r)$.

In Fig. 2 we present the vacancy dispersion relation which is analyzed within the tight-binding approximation of Ref. [23], obtaining tunneling amplitudes $t_z = 0.45(5)$ K and $t_\perp = 0.50(5)$ K and effective masses $m_z^* = 0.45(5)$ and $m_\perp^* = 0.42(5)$ in units of the bare ^{4}He mass. We find that the effective mass is about half that of ^{4}He atoms, and nearly isotropic [24]. There is a small difference with the variational calculation of Ref. [23] which reported $m_\perp^* = 0.31$ in the basal plane and $m_z^* = 0.38$ along the ΓA direction.

In Fig. 3 we show the vacancy-vacancy correlation function, decreasing exponentially with distance. This proves that interactions between vacancies are attractive and strong enough to form a bound state. From the exponential fit we estimate the binding energy to be $E_b = 1.4(5)$ K. We conclude that the hcp structure at n_c is kinetically unstable against collapse into the liquid state, and a supersolid in a decompressed crystal is not possible in the homogeneous setup.

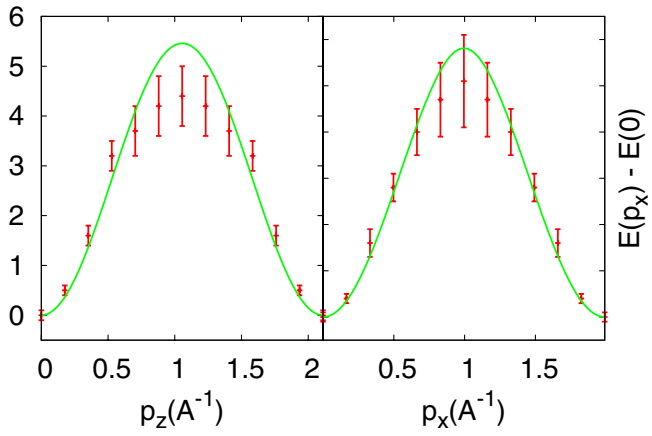

FIG. 2 (color online). Vacancy dispersion relation of the lowest band along the reciprocal lattice directions p_z and p_x for a system of $12 \times 12 \times 12$ particles calculated at a density $n = 0.0292$ Å^{-3} and temperature $T = 0.2$ K. For the hcp lattice with two atoms in the unit cell there are two hopping amplitudes, one in the basal plane ($t_\perp$), and one (t_z), along the ΓA direction. At low temperatures the Monte Carlo method will project out the lowest branch of the dispersion relation.

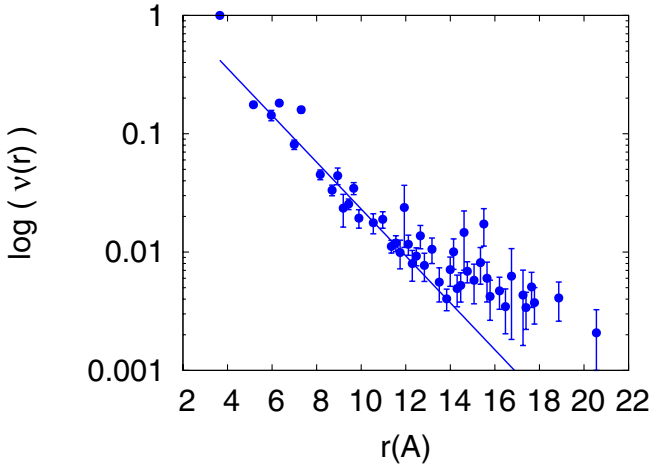

FIG. 3 (color online). The probability of finding two vacancies at a distance r. The initial decay is fitted with the exponential function $\nu(r) \propto \exp(-2\sqrt{m^* E_b} r)$ with $E_b = 1.4(5)$ K. The simulation was performed for the hcp solid with $6 \times 6 \times 6$ lattice points and 214 atoms at a density $n = 0.0292$ Å^{-3} and temperature $T = 0.5$ K. At large distances the probability reaches a plateau due to finite temperature effects.

Anisotropic diagonal traceless strain.—We now turn to the study of crystals with nonzero diagonal components of the strain tensor u_{xx}, u_{yy}, and $u_{zz} = -u_{xx} - u_{yy}$. The phenomenology of the strain-induced supersolidity in the hcp structure [18] is based on the minimal energy density allowed by symmetry: $\epsilon = [a_z u_{zz} + a_\perp(u_{xx} + u_{yy})]|\psi|^2$, where ψ is the superfluid order parameter and a_z, $a_\perp$ are some constants. In the simulations we find (see Fig. 4) that a strain $u_{zz} = -2u_{xx} = -2u_{yy}$ of about 10–12% is necessary to close the gap for vacancy formation, and at higher strain the hcp structure collapses. Such a strain corresponds to a stress $\sigma_{zz} = C_{33}u_{zz} + C_{13}(u_{xx} + u_{yy}) = (C_{33} - C_{13})u_{zz}$ of approximately 50 bar, hardly achievable under realistic experimental conditions. Our data show that, within error bars, there is initially no dependence of the gap on anisotropic compression. Thus, the linear coupling to the anisotropic strain is close to zero, $a_z \approx a_\perp$, and one has to go beyond linear theory to account for the observed effects, including the closing of the gap.

Shear stress.—By symmetry there is no linear coupling between $|\psi|^2$ and shear strain characterized by the off-diagonal components u_{zx}, u_{yx}, u_{zy}. The anticipated strain dependence of the gap is quadratic, $\Delta_V = \Delta_V^{(0)}[1 - (u_{zx}^2 + u_{zy}^2)/u_c^2]$. Our results in Fig. 5 allow us to estimate the critical value of shear strain as $u_c \approx 0.15$. Using measured values of the elastic modulus $C_{44} \approx 120$–130 bar [25] the corresponding critical shear stress, $2C_{44}u_c$, for closing the gap in the hcp ^{4}He is about 35 bar.

Superfluidity along crystalline defects.—Even though the homogeneous strain-induced supersolid phase is unstable, it can form locally if nonuniform strain close to structural defects exceeds the critical value and destabilizes parts of the crystal. Candidates for such highly strained superfluid defects include dislocations [17–19], with edge dislocations attracting most attention in the past because they produce strain linearly coupled to the superfluid order parameter. Contrary to expectations, the first numerical evidence for superfluidity in the dislocation core was reported for screw dislocations oriented along the $\hat{z}$ direction [26]. These are characterized by nonzero values of u_{zx} which can be estimated by dividing the modulus of the Burgers vector $b_z = \sqrt{8/3}a$ by twice the circumference of the circle going through the atoms closest to the core, $4\pi a/\sqrt{3}$. The estimated strain [27] $\sqrt{u_{xz}^2 + u_{yz}^2} = 1/(\sqrt{2}\pi) \approx 0.22$ exceeds the threshold value of $u_c = 0.15$ found above for shear stress, explaining why the superfluid density of the screw dislocation involves nearly all atoms closest to the nucleus.

Dislocations in solid ^{4}He (with two atoms per unit cell) may optimize energy by splitting the core and thus halving the Burgers vector and the strain. We observe such splitting in ongoing simulations [28] for the edge dislocation with core along the $\hat{y}$ direction and Burgers vector along $\hat{z}$. Here the half-Burgers vector is $\sqrt{2/3}a$. The estimated strain [27] is $u_{\rm def} \approx 0.13$, marginally larger than the critical strain of $u_c \approx 0.12$ in Fig. 4. We expect this type of dislocation to be (weakly) superfluid, as is confirmed by simulations [28]. For the split-core edge dislocation along $\hat{y}$ with Burgers vector along $\hat{x}$, the strain is $u_{\rm def} \approx 0.08$ and below the threshold value. Direct simulations show that this type of defect is insulating [28]. The grain boundary with low tilting angle can be represented by the edge dislocation sheet, and its superfluidity (or absence thereof) is consis-

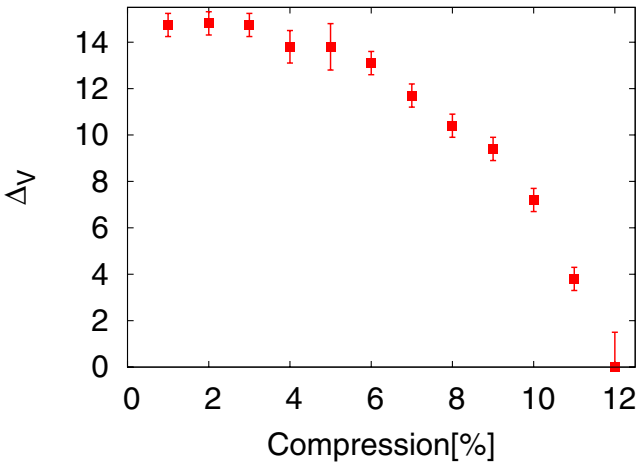

FIG. 4 (color online). Gaps for vacancy formation under anisotropic diagonal traceless strain. An ideal hcp solid of size $6 \times 8 \times 8$ at density $n = 0.0292$ Å^{-3} is subjected to anisotropic compression along the z direction. The volume is kept constant by dilatation in the basal plane. The temperature is $T = 0.25$ K.

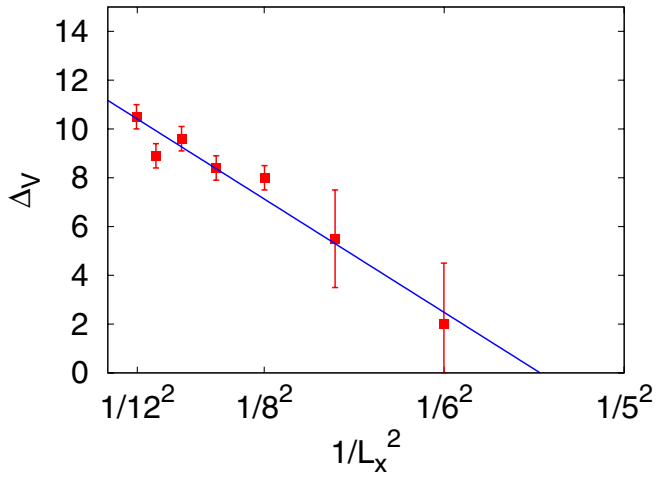

FIG. 5 (color online). Gaps for vacancy formation under shear stress. An ideal hcp solid of size $L_x \times 12 \times 12$ at density $n = 0.0292$ Å^{-3} is subjected to shear stress along the z direction by uniformly deforming the sample so that the atoms at boundary $x = aL_x$ (with $a = 3.645$ Å) are shifted exactly by one lattice period $c = \sqrt{8/3}a$ along the c axis in order to match the periodic boundary conditions. The strain introduced is defined by $u_{zx} = \sqrt{2/3}/L_x$ and, for the chosen sizes $L_x = 12$–5, the strain ranges from 0.07 down to 0.15. The temperature is $T = 0.2$ K.

tent with our earlier observations [29]. The mechanism of splitting the core is not effective for screw dislocations.

Summarizing, there is growing experimental evidence for unexpected properties of solid ^{4}He at low temperature, including mass superflow through the solid. The observed dependence on sample history, growth conditions, annealing, and cooling procedures indicate that crystalline defects are important for our understanding of the most quantum solid in nature. The actual structure of defects is essentially unexplored territory, not less interesting than the solid matrix they reside in. We determined the critical values of the strain which are required to destabilize the hcp structure of ^{4}He by closing its insulating gap, and find that these thresholds are small enough to be exceeded at the dislocation cores and grain boundaries.

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[1] A. F. Andreev and I. M. Lifshitz, Sov. Phys. JETP **29**, 1107 (1969).
[2] G. V. Chester, Phys. Rev. A **2**, 256 (1970).
[3] N. Prokof'ev and B. Svistunov, Phys. Rev. Lett. **94**, 155302 (2005).
[4] M. W. Meisel, Physica (Amsterdam) **178B**, 121 (1992).
[5] E. Kim and M. H. W. Chan, Nature (London) **427**, 225 (2004); Science **305**, 1941 (2004).
[6] A. S. C. Rittner and J. D. Reppy, Phys. Rev. Lett. **97**, 165301 (2006); **98**, 175302 (2007).
[7] Y. Aoki, J. C. Graves, and H. Kojima, Phys. Rev. Lett. **99**, 015301 (2007).
[8] M. Kondo, S. Takada, Y. Shibayama, and K. Shirahama, J. Low Temp. Phys. **148**, 695 (2007).
[9] A. Penzev, Y. Yasuta, and M. Kubota, J. Low Temp. Phys. **148**, 677 (2007).
[10] S. Balibar and F. Caupin, J. Phys. Condens. Matter **20**, 173201 (2008).
[11] M. Boninsegni, N. Prokof'ev, and B. Svistunov, Phys. Rev. Lett. **96**, 105301 (2006).
[12] B. K. Clark and D. M. Ceperley, Phys. Rev. Lett. **96**, 105302 (2006).
[13] D. S. Greywall, Phys. Rev. B **16**, 1291 (1977).
[14] J. Day, and J. Beamish, Phys. Rev. Lett. **96**, 105304 (2006).
[15] S. Sasaki, R. Ishiguro, F. Caupin, H. J. Maris, and S. Balibar, Science **313**, 1098 (2006).
[16] M. W. Ray and R. B. Hallock, Phys. Rev. Lett. **100**, 235301 (2008);(private communications).
[17] S. I. Shevchenko, Sov. J. Low Temp. Phys. **13**, 61 (1987).
[18] A. T. Dorsey, P. M. Goldbart, and J. Toner, Phys. Rev. Lett. **96**, 055301 (2006).
[19] J. Toner, Phys. Rev. Lett. **100**, 035302 (2008).
[20] M. Boninsegni, N. Prokof'ev, and B. Svistunov, Phys. Rev. Lett. **96**, 070601 (2006); Phys. Rev. E **74**, 036701 (2006).
[21] M. Boninsegni, A. B. Kuklov, L. Pollet, N. V. Prokof'ev, B. V. Svistunov, and M. Troyer, Phys. Rev. Lett. **97**, 080401 (2006).
[22] J. F. Jarvis, D. Ramm, and H. Meyer, Phys. Rev. **170**, 320 (1968).
[23] D. E. Galli and L. Reatto, Phys. Rev. Lett. **90**, 175301 (2003).
[24] As a side remark we note that the equilibrium density of vacancies at finite temperature $n_V = (m^* T/2\pi)^{3/2} \times e^{-\Delta_V/T} \approx 0.015 n e^{-\Delta_V/T}$ involves a very small preexponential factor in addition to the exponential suppression, which rules out their possible role in thermodynamics at temperatures below 1 K.
[25] D. S. Greywall, Phys. Rev. B **16**, 5127 (1977).
[26] M. Boninsegni, A. B. Kuklov, L. Pollet, N. V. Prokof'ev, B. V. Svistunov, and M. Troyer, Phys. Rev. Lett. **99**, 035301 (2007).
[27] A. M. Kosevich, E. M. Lifshitz, L. D. Landau, and L. P. Pitaevskii, *Theory of Elasticity*, Theoretical Physics Vol 7 (Reed Educational and Professional Publishing, Ltd., Oxford, U.K., 1986), 3rd ed.
[28] S. G. Söyler *et al.* (unpublished).
[29] L. Pollet *et al.*, Phys. Rev. Lett. **98**, 135301 (2007).