# Superfluidity of Grain Boundaries in Solid $^4$He

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By large-scale quantum Monte Carlo simulations we show that grain boundaries in $^4$He crystals are generically superfluid at low temperature, with a transition temperature of the order of $\sim$0.5 K at the melting pressure; nonsuperfluid grain boundaries are found only for special orientations of the grains. We also find that close vicinity to the melting line is not a necessary condition for superfluid grain boundaries, and a grain boundary in direct contact with the superfluid liquid at the melting curve is found to be mechanically stable and the grain-boundary superfluidity observed by Sasaki *et al.* [Science **313**, 1098 (2006)] is not just a crack filled with superfluid.

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Superfluid grain boundaries (gb) were proposed as a plausible scenario [1–3] to explain the effect of nonclassical rotational inertia (NCRI) in solid $^4$He discovered by Kim and Chan [4]. An observation by Rittner and Reppy [5] that the NCRI signal can be eliminated through annealing was the first explicit evidence that crystalline defects are of crucial importance. The remarkable direct experimental observation of grain-boundary superfluidity (at the melting point) by Sasaki *et al.* [6] confirms the early theoretical prediction and marks the beginning of a new stage in the study of the supersolid phase of helium.

In this Letter we expand on our previously reported preliminary results [3] and show that a grain boundary in solid helium is generically superfluid at low temperatures. The transition temperature, $T_c$, is strongly dependent on the crystallite orientation: while it is typically of the order of $\sim$0.5 K, grain boundaries with special relative orientations of the two grains are found to be insulating (nonsuperfluid). We also obtain strong evidence that a grain boundary in contact with the superfluid liquid, as shown in Fig. 1, is mechanically stable.

This latter question is important for the interpretation of the experimental observation of superflow in crystals with grain boundaries [6]. Since these experiments are carried out under the conditions of phase coexistence between a crystal and a liquid, the observed effect could either be true superflow along a grain boundary, or rather a thin liquid-filled crack in a crystal. The answer depends on the relationship between the three surface tensions, $\sigma_1$, $\sigma_2$, and $\sigma_{\rm gb}$ (see Fig. 1). Mechanical and energetic stability of the grain boundary require that

$$\sigma_{\rm gb} < \sigma_1 + \sigma_2. \tag{1}$$

If this inequality is not satisfied, the liquid penetrates between the two crystallites, and a crack is formed. We will show that this configuration is stable. The grain-boundary superfluidity observed by Sasaki *et al.* is thus not merely the manifestation of a crack in the crystal filled with liquid.

Our path integral Monte Carlo (PIMC) simulations are based on the continuous-space worm algorithm [7]. For spatial imaging, we employ two slightly different techniques. The first consists of producing condensate maps, which are maps of the condensate wave function, in thin slices of our sample. Within the worm algorithm, this is accomplished by recording spatial positions of the two open ends of the worms, when they are sufficiently far away from each other such that correlations between them are negligible [8]. As a result, the density of points in the map is proportional to the condensate wave function. The second technique is based on the winding-cycle maps. Here, we collect statistics of instantaneous particle positions, by considering only particles which participate in macroscopic exchange cycles characterized by nonzero winding numbers, i.e., cycles that directly contribute to the superfluid response [9].

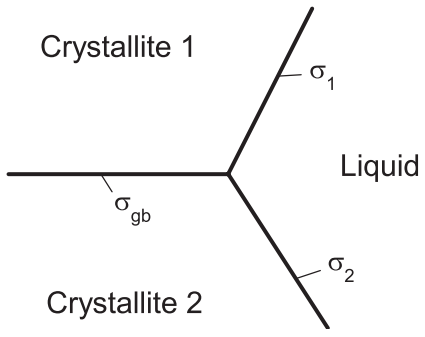


FIG. 1. Sketch of the equilibrium configuration of two crystallites in contact with liquid. The configuration is mechanically stable only if the condition (1) is met.

 

*Stability of the grain-boundary-liquid junction.*—In order to check whether a grain boundary is destroyed when brought into contact with the liquid, we perform a direct simulation of two crystallites in contact with liquid sketched in Fig. 1. Our simulation setup, shown in Fig. 2, consists of two truncated solid pyramids with random crystalline orientation are placed on top of each other. The rest of the volume is filled with liquid.

Since the goal of this particular simulation is to study the stability of a grain boundary we can work at a relatively high temperature, $T = 0.8$ K, which significantly enhances the performance of our algorithm: the size of the configuration in the imaginary-time direction is reduced, and the relaxation of the liquid-solid and solid-solid interfaces is numerically facilitated in the Monte Carlo simulations [10]. The simulation is carried out in the grand canonical ensemble, with the chemical potential fixed at the phase coexistence point. The equilibrium number of atoms in our sample was measured to be about 13 660. In order to stabilize the solid phase in the system, we pin (i.e., do not update) solid atoms in the vicinity of the pyramid bases.

In Fig. 3 we show the condensate map of the sample. We see that the grain boundary between the two crystallites is a robust quantum object with a thickness of order 3 (see also Fig. 5) and did not disappear during the simulation run. The system has converged to a state where the liquid and solid phase coexist. Compared to the initial configuration, the shapes of the crystallites (including the angles) have noticeably changed. This is not surprising since the optimal shape of the crystal-liquid interface depends on the particular orientation of the crystallite axes with respect to the interface.

*Superfluidity of grain boundaries.*—Having confirmed the stability and superfluidity of grain boundaries in a generic sample, we next perform a more systematic study of the properties of grain boundaries, extending our previously reported simulations [3], where the superfluidity had been observed, but only for one particular polycrystalline structure, and grain-boundary superfluidity could not clearly be distinguished from grain edge superfluidity.

The simulation of a grain boundary between two truncated pyramids with basal length $L = 24$ is computationally very expensive since it requires ~13 660 atoms, and is by far the largest simulation of solid helium performed to date. Reducing the size of the pyramids is not an option, since the typical width of the superfluid grain-boundary region, estimated from Fig. 3 (see also Fig. 5), is ~3. Hence, in the two-pyramid samples of linear sizes significantly smaller than $L = 20$–$30$, the grain boundary is not well defined, and the system is not supposed to be metastable. For instance, in a sample with $L = 12$, we found that the grain boundary and the two crystallites (apart from the pinned atoms) melt.

To study the properties of generic grain boundaries we use the sample geometry shown in Fig. 4, consisting of two equally sized cuboids placed on top of each other.

An important result of our simulations is that, as already observed in our preliminary studies [3], not all grain boundaries in the hcp $^4$He crystal are superfluid. Grain boundaries featuring an extra symmetry such as stacking

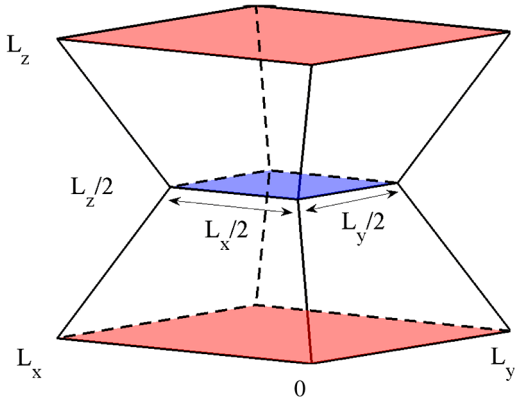


FIG. 2 (color online). Initial setup for the simulation of a grain boundary in contact with a liquid. Two truncated pyramids are placed on top of each other. The basal plane of both pyramids is a square with size $L_x \times L_y = L \times L$, with $L = 24$ [12]. The upper and lower pyramid have different random orientations and the height of both pyramids is $L_z = L/2$. The upper facets of the truncated pyramids are squares of initial size $L/2 \times L/2$, and form a grain boundary between the two crystallites, indicated by the blue square. Liquid fills the volume outside the crystallites. Periodic boundaries are used in the $x$ and $y$ directions, while atoms in the $z = 0$ and $z = L$ plane, drawn in red, are pinned. See the auxiliary material for more details [13].

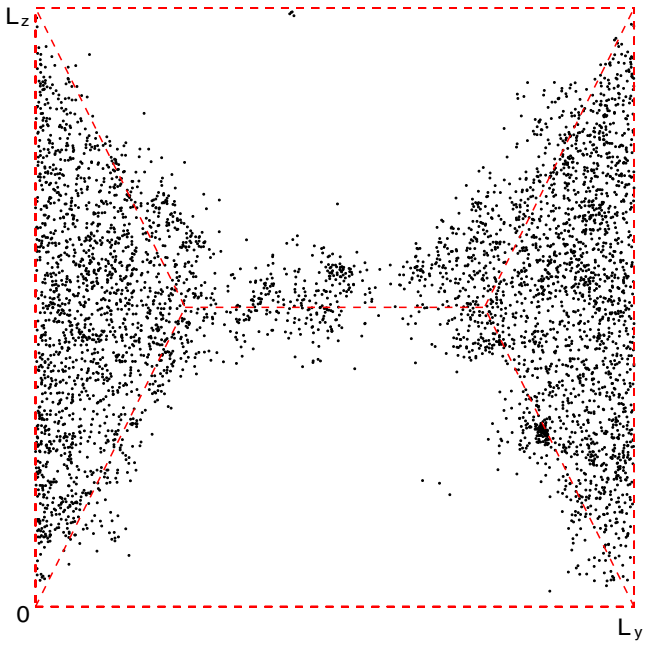


FIG. 3 (color online). Condensate map of the two-pyramid sample. The map represents (by the density of points) the condensate wave function in the slice $x \in [0.4L, 0.6L]$ averaged over the $x$ direction. The initial setup is shown by dashed lines.

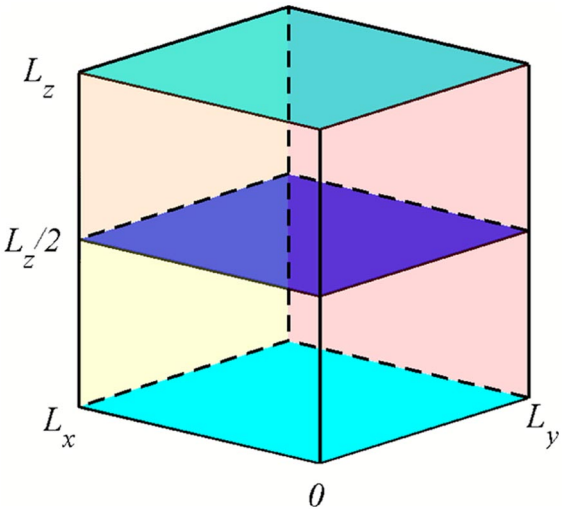


FIG. 4 (color online). Sketch of the initial sample of two cuboid crystallites with different random orientations (see Ref. [13] for more details) placed on top of each other. The basal plane of both cuboids is a square with size $L_x \times L_y = L \times L$, with $L = 12$. The height of both cuboids is $Lz/2 = 7$, yielding a grain boundary in the $z = 0$ plane (light blue) and a grain boundary in the $z = Lz/2$ plane (dark blue). Motion across the $xz$ boundary was suppressed by pinning all atoms at a distance smaller than 0.75 from the $y = 0 (y = L_y)$ boundary indicated in red. This ensures that any superfluid response in the $x$-direction is due to the superfluidity of two horizontal grain boundaries [14]. An additional grain boundary due to periodic boundary conditions arises for each crystallite at the $x = 0 (x = L_x)$ plane (yellow and orange for the lower and the upper cuboid, respectively), but these do not affect the superfluid response in the $x$ direction.

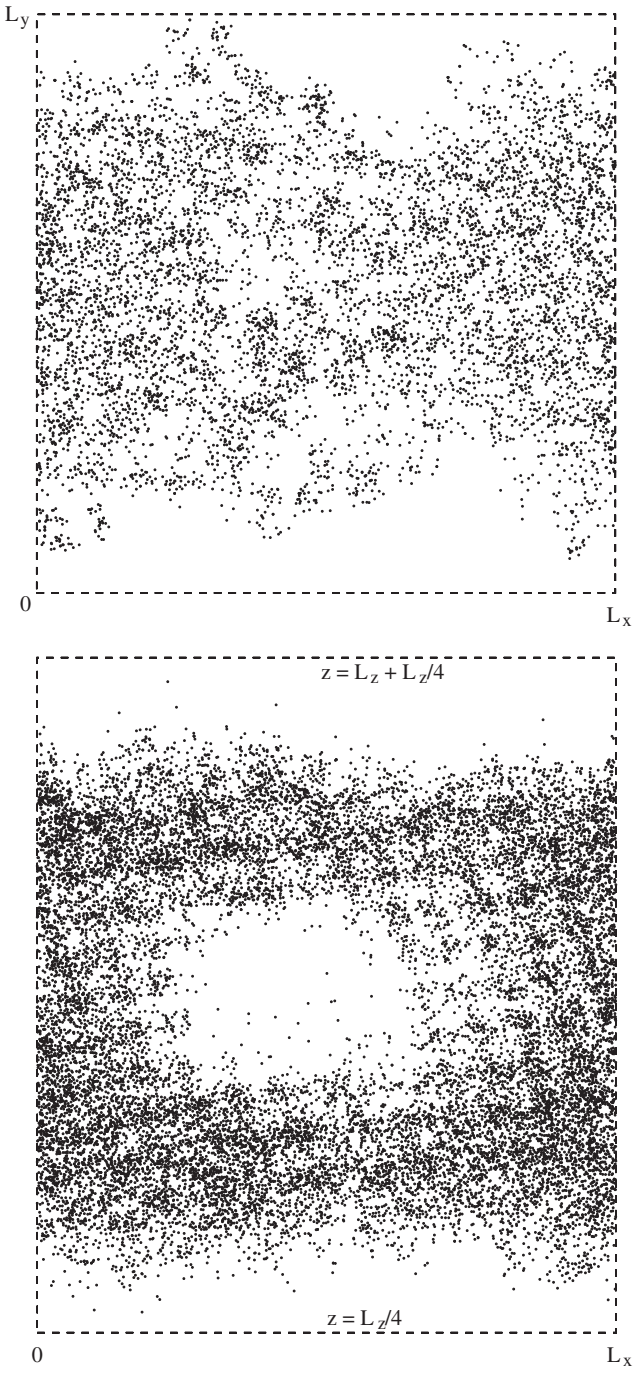


FIG. 5. Phase-coherence properties (winding-cycles maps) of grain boundaries in the $^4$He sample shown in Fig. 4. Upper panel: projection on the $xy$ plane of the data for the upper half of the sample containing one of the two grain boundaries. Lower panel: ($z$-shifted) projection of all the data points on the $xz$ plane. Note that the $x = 0$ ($x = L_x$) grain boundary (induced by boundary conditions) of one of the two crystallites turned out to be insulating (see Fig. 6).

faults and special grain boundaries with nicely matching angles similar to the one shown in Fig. 6, are insulating.

The generic case, however, are superfluid grain boundaries. Results of a typical simulation at $T = 0.25$ K, 2000 particles and $n = 0.0287$ Å$^{-3}$ (melting density) are shown in Fig. 5. The winding-cycle map clearly reveals superfluidity along the $x$ direction. Based on the relatively large system size utilized in this study, we argue that our observed superfluid signal reflects macroscopic grain-boundary superfluidity, and is not an artifact, due, for example, to the vicinity of a superfluid ridge—the intersection of the grain boundary of interest and the additional grain boundary caused by periodic boundary conditions at $x = 0$ ($x = L$). Incidentally, we note that in the area close to the ridge, of size $\sim 3$, the density of the map is significantly increased, which is consistent with our general observation that nearly all ridges have robust phase-coherence properties.

An interesting observation is that the density in the vicinity of superfluid grain boundaries is close to that of the crystal, which again confirms that the superflow along the grain boundary is not a liquid-filled crack.

On the basis of a number of simulations similar to the one presented in Fig. 5, performed at different pressures, temperatures, and crystallite axes orientations, we conclude that generic grain boundaries are superfluid with typical (orientation dependent) transition temperatures of about half a kelvin. The width of the superfluid grain-

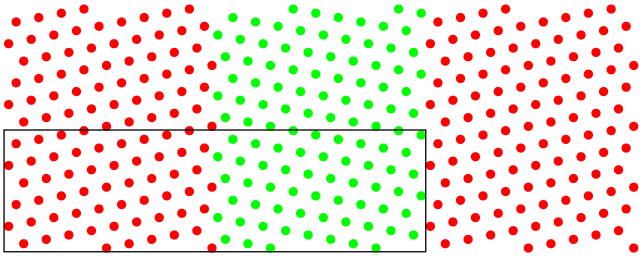

FIG. 6 (color online). The base plane of an insulating grain boundary between two crystallites that differ only by a rotation about the axis perpendicular to the base plane. Outside the box we show the periodic continuation of the sample. Even with this moderate system size, it is clearly established that the grain boundary is not superfluid. Decreasing the angle between the two crystallites strengthens the insulating character, due to the better match between the atoms of different crystallites. Since this grain boundary is insulating and can be viewed as a wall of sparsely spaced edge dislocations, we conclude that edge dislocations along the $c$ axis are insulating as well.

boundary region is $\sim 3$. We conjecture that the maximum possible $T_c$ for grain boundary should be at least smaller than the transition temperature of the overpressurized liquid of the same density as the crystal. By simulating superfluid properties of the overpressurized liquid, we found the transition temperature at the density $n = 0.0287$ Å$^{-3}$ to be 1.5(1) K. We thus take this value as the upper bound for grain-boundary $T_c$ at the melting pressure.

In order to provide a further assessment of the robustness of our conclusions, we have repeated the same study, replacing helium with molecular *para*-hydrogen, at the same low temperatures. In this case, individual particles have a mass which is half that of helium atoms, whereas the interaction potential is approximately 3 times deeper. No evidence of superfluidity was ever observed in this case, in agreement with experimental findings [11].

Summarizing, based on a direct quantum Monte Carlo simulation of a grain boundary in hcp $^4$He in contact with liquid under the conditions of phase coexistence, we argue that it is thermodynamically stable against dissolution into two crystallites separated by a crack. This lends theoretical support to the observation of Sasaki *et al.* [6] of generic superfluid grain boundaries. We have studied superfluid properties of grain boundaries and found that special grain boundaries of higher symmetry are insulators. On the other hand, grain boundaries of a general form are found to be superfluid, with typical transition temperatures of the order of $\sim 0.5$ K. The relevance of these findings to the interpretation of the experiment of Kim and Chan may be assessed once the concentration of grain boundaries in the experimental samples is measured.

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[1] N. Prokof'ev and B. Svistunov, Phys. Rev. Lett. **94**, 155302 (2005).
[2] E. Burovski, E. Kozik, A. Kuklov, N. Prokofev, and B. Svistunov, Phys. Rev. Lett. **94**, 165301 (2005).
[3] Our preliminary results on superfluidity of grain boundaries in $^4$He were reported at the KITP Miniprogram: The Supersolid State of Matter, 6-17 February 2006, http://online.kitp.ucsb.edu/online/smatter_m06/svistunov.
[4] E. Kim and M. H. W. Chan, Nature (London) **427**, 225 (2004); Science **305**, 1941 (2004).
[5] A. S. C. Rittner and J. D. Reppy, Phys. Rev. Lett. **97**, 165301 (2006).
[6] S. Sasaki, R. Ishiguro, F. Caupin, H. J. Maris, and S. Balibar, Science **313**, 1098 (2006).
[7] M. Boninsegni, N. Prokof'ev, and B. Svistunov, Phys. Rev. Lett. **96**, 070601 (2006); Phys. Rev. E **74**, 036701 (2006).
[8] M. Boninsegni, N. V. Prokof'ev, and B. V. Svistunov, Phys. Rev. Lett. **96**, 105301 (2006).
[9] E. L. Pollock and D. M. Ceperley, Phys. Rev. B **36**, 8343 (1987).
[10] In real-time dynamics the mobility of the interface is, however, slower; see S. Balibar, H. Alles, and A. Y. Parshin, Rev. Mod. Phys. **77**, 317 (2005).
[11] A. C. Clark, X. Lin, and M. H. W. Chan, Phys. Rev. Lett. **97**, 245301 (2006).
[12] All distances are measured in units of the mean interparticle distance in the solid phase, $n^{-1/3}$, at the $T = 0$ melting density of a $^4$He hcp crystal, $n = 0.0287$ Å$^{-3}$.
[13] See EPAPS Document No. E-PRLTAO-98-075714 for a detailed explanation of the preparation of the initial sample. For more information on EPAPS, see http://www.aip.org/pubservs/epaps.html.
[14] Otherwise, we would have to deal with the "shunting" superfluid response of grain boundaries arising, by periodicity, within the same crystallite at $y = 0 (y = L_y)$ interfaces.