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M Boninsegni

Nikolai Prokof‘ev
University of Massachusetts - Amherst, prokofev@physics.umass.edu

Boris Svistunov
University of Massachusetts - Amherst, svistunov@physics.umass.edu

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Superglass Phase of $^4$He

Massimo Boninsegni$^1$, Nikolay Prokof'ev$^{2,3,4}$, and Boris Svistunov$^{2,3}$

$^1$Department of Physics, University of Alberta, Edmonton, Alberta T6G 2J1
$^2$Department of Physics, University of Massachusetts, Amherst, MA 01003
$^3$Russian Research Center “Kurchatov Institute”, 123182 Moscow
$^4$Department of Physics, Cornell University, Ithaca, NY 14850

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We study different solid phases of $^4$He, by means of Path Integral Monte Carlo simulations based on a recently developed worm algorithm. Our study includes simulations that start off from a high-$T$ gas phase, which is then “quenched” down to $T=0.2$ K. The low-$T$ properties of the system crucially depend on the initial state. While an ideal hcp crystal is a clear-cut insulator, the disordered system freezes into a superglass, i.e., a metastable amorphous solid featuring off-diagonal long-range order and superfluidity.

The remarkable observation by Kim and Chan of a non-classical moment of inertia in solid $^4$He has generated a new wave of interest in the possible superfluid phase of a solid. Supersolidity of $^4$He is still contro- versial, both at the experimental and theoretical levels. Two of us have recently proven that, irrespective of its microscopic structure, any supersolid crystal should contain gapless vacancies and/or interstitials. In other words, any continuous-space supersolid is generically incommensurate (i.e., the number of atoms per unit cell is not an integer) and squeezeable, i.e., by applying pressure it should be possible to squeeze matter from a container with supersolid into a buffer volume containing the same supersolid. However, this very experiment has yielded a negative result for solid $^4$He.

A wealth of numerical studies clearly indicate that $^4$He is a commensurate (thus insulating) crystal. The finite activation energy of a vacancy computed numerically is large, ~ 15 K, and claimed consistent with the experimental observations. The activation energy of an interstitial, ~ 50 K, is significantly larger than that of a vacancy. A simulation study of exchanges in an ideal hcp crystal, yielded indirect evidence that the system is not superfluid. In sharp contrast, the variational ($T=0$) calculation of Ref. claims a finite condensate fraction in the commensurate $^4$He crystal. Thus, additional investigation is warranted.

The experiment of Kim and Chan itself has revealed a number of facts pointing to a strongly inhomogeneous scenario of superfluidity, chiefly the contaminating effect of a small concentration of $^3$He, and non-XY behavior of the superfluid density at the critical temperature. The need of exploring inhomogeneous (metastable) scenarios of supersolidity, dictated both by theory and experiments, has already resulted in some relevant theoretical developments, revealing superfluid interfaces in a lattice solid and a superfluid layer at the boundary between the $^4$He crystal and a disordered substrate.

The numerical observation of a metastable disordered supersolid, (a superglass phase of $^4$He) is reported in the present Letter. To be specific in the definition, by glass we mean a spatially disordered (metastable) phase, indistinguishable from a solid on a time scale much shorter than the typical relaxation time, $t_{rel}$, which in turn should be dramatically longer than the inverse Debye frequency, $\omega_D^{-1}$. Superglass is the term that we use for such a phase, if it also displays superfluidity. Note that our definition of glass does not address the behavior of the system at time scales $t \gtrsim t_{rel}$, whereupon it may undergo structural relaxation into the polycrystalline sample, or simply behave as a very viscous liquid.

In Ref. 10, the idea of glassification of overpressurized liquid $^3$He was put forward, in order to explain a striking experimental outcome, i.e., the absence of bulk solid nucleation under fast (about 1 µs) acoustic wave compression pulses, up to pressures as high as ~ 160 bar. The authors conjectured that the glassy phase is normal (though the experiment was done at $T = 0.05$ K and the adiabatic heating was estimated to be below 0.1 K); the absence of superflow towards the nucleus center would dramatically suppress the rate of growth of the crystal. Conceptually, the finding of the present Letter is different, but we believe relevant to the interpretation of the experiment of Ref. 10. Jamming of structural relaxation does not per se exclude superfluidity. Crystallization is suppressed by the mere fact that the normal component forms a glassy solid, implying that further evolution towards a lower-energy polycrystal structure necessarily involves a chain of exponentially rare quantum-tunnelling or thermoactivation events, rather than a rapid growth of the supercritical nucleus. Indeed, the boundary between the perfect-crystal nucleus and the superglass, is a solid-solid interface which realizes a pronounced local energy minimum. Its evolution should therefore imply either quantum tunnelling (in the $T \to 0$ limit), or thermoactivation.

Our study is based on accurate Path Integral Monte Carlo (PIMC) simulations of condensed $^4$He, making
use of a recently developed worm algorithm \cite{11}. This method allows for efficient sampling and accurate determination of the single-particle Green function and superfluid density, for systems comprising a relatively large number \( N \) of particles (of the order of several thousand). Specifically, we address the following two issues: (i) Is it possible to obtain definitive first-principle theoretical evidence that an ideal \( \text{hcp} \) \(^4\text{He} \) crystal is an insulator? (ii) What happens to a sample of liquid \(^4\text{He} \) quenched through the first-order liquid-solid phase transition?

We consider a system of \( N \) \(^4\text{He} \) atoms (\( N=216 \) and 800), at a temperature 0.2 K \( \leq T \leq 1 \) K, and at the two densities \( n=0.0292 \) (0.0359) \( \text{Å}^{-3} \), corresponding to an ideal \( \text{hcp} \) \(^4\text{He} \) crystal at a pressure of approximately 32 (155) bars \cite{12}. The sample cell geometry with periodic boundary conditions is designed to fit an ideal \( \text{hcp} \) crystal. We use the standard microscopic model of \(^4\text{He} \), based on the Aziz pair potential \cite{13}.

In Figs. 1 and 2 we show data for the pair correlation function \( g(r) \) and the single-particle density matrix \( n(r) \). For both the near-melting density of \( n=0.0292 \text{ Å}^{-3} \) and the higher density of \( n=0.0359 \text{ Å}^{-3} \) we study two samples, differing in one respect only, namely their initial configurations before equilibration.

The single-particle density matrix is defined as \( n(r, r') = \langle \psi^\dagger (r) \psi (r') \rangle \) where \( \psi (r) \) is the particle annihilation operator, \( \hat{\rho} (r) = \hat{\psi}^\dagger (r) \hat{\psi} (r) \) is the local \(^4\text{He} \) density operator, and \( (...) \) stands for thermal average. It is customary to display the spherically averaged function

\[ n(r) = \frac{1}{4\pi r^2} \int d\Omega \int d^3r' \ n(r', r'+r) \]  

(1)

where \( V \) is the volume of the system. This is the quantity shown in Fig. 2.

When the simulation is started from an initial configuration corresponding to an ideal \( \text{hcp} \) crystal, we consistently find an exponential decay of \( n(r) \) at large distances, with short-range oscillations due to coordination-sphere effects. We observe no change in the results between the temperatures of 0.2 and 1 K, to indicate that those shown in Fig. 2 are essentially ground state estimates. This result provides a robust confirmation that an ideal \( \text{hcp} \) crystal is not a Bose condensate (superfluid).

This conclusion is consistent with the theoretical expectation that a crystal with finite activation energies for vacancies and interstitials will not display superfluidity \cite{2}, and is in agreement with arguments based on the statistics of exchange cycles observed in the same system \cite{6}. The results and conclusions of Ref. \cite{6} appear therefore to be erroneous, possibly artifacts of the variational approach.

At present, there is no clear understanding of what crystalline defects dominate in the experimental samples of Ref. \cite{1}. It is not known whether individual dislocations, dislocation sheets and networks, or grain boundaries in bulk \(^4\text{He} \) may underlie the experimentally observed superfluid response, though model simulations of domain walls in quantum solids hint at such possibilities \cite{7}. But regardless of their nature, in the absence of crystalline defects no theoretical interpretation seems viable of the experiments reported in Ref. \cite{1} in terms of superfluid response.

In order to investigate scenarios of broken translation invariance not involving a perfect crystal (though perhaps not directly related to the experiment of Ref. \cite{1}), we designed a simulation protocol aimed at mimicking a “quenching” experiment (namely one in which liquid Helium is suddenly, rapidly cooled) obviously mak-
ing allowance for the important differences between the imaginary-time PIMC dynamics and the real-time dynamics of actual physical systems (see discussion below). Starting from an initial configuration characteristic of a high-\(T\) gas phase, we “quench” the system down to the temperature \(T = 0.2\) K by \(> 10^4\) PIMC sweeps. One “sweep” is defined as the number of accepted updates sufficient to sample the entire Path Integral configuration. We then run the simulation long enough to achieve stability of statistical averages for structural properties and for the single-particle density matrix.

The phase that emerges from disorder resembles the \(hcp\) crystal only at short interatomic distances (of about two-three coordination spheres), with no diagonal long-range order (see Fig. 1). Moreover, this phase has a condensate fraction \(n_c = 0.5\%\) (see Fig. 2), and a surprisingly large superfluid fraction \(\rho_s = 0.6(1)\) at \(n = 0.0292\) \(\text{Å}^{-3}\) and \(\rho_s = 0.07(2)\) at \(n = 0.0359\) \(\text{Å}^{-3}\). Though the superfluid fraction is strongly suppressed with pressure the condensate fraction is reduced by merely 50%.

The important observation is that \(^4\text{He}\) can remain in the metastable superfluid state at solid-state densities, even at fairly high pressures. This observation is consistent with a previous study [14], predicting overpressurized liquid \(^4\text{He}\) to remain superfluid (at \(T = 0\)) to arbitrarily high density. The nature of the superfluid phase will generally depend on pressure, temperature, and the experimental time scale. For example, one may expect that the lower density finite-temperature phase should be just a superfluid, but with a rather viscous normal component. On the other hand, with increasing density such a normal component may evolve into a glass, with a diverging (i.e., unobservably large) viscosity.

In order to study whether and how the system breaks translation symmetry, we calculate the condensate wave function \(\phi(\mathbf{r})\). The worm algorithm offers direct access to the one-body density matrix which in the presence of off-diagonal long-range order factorizes at large separation \(|\mathbf{r} - \mathbf{r}'|\)

\[
\langle \psi(\mathbf{r}) \psi(\mathbf{r}') \rangle \rightarrow \phi(\mathbf{r}) \phi(\mathbf{r}')
\]

In Fig. 3 we show two-dimensional \(xy\)-maps of the condensate wave function \(\phi(x, y, z)\) at \(n = 0.0359\) \(\text{Å}^{-3}\), for ten (equally spaced) slices along the \(z\)-axis. The data shown in Fig. 3 represent long simulation-time averages, not instantaneous snapshots. Aside from the obvious observation that the system has manifestly broken translational invariance, the results suggest no obvious interpretation of the disordered pattern for \(\phi(\mathbf{r})\) in terms of dislocations or grain boundaries [15]. Correlations barely extend over two slices in Fig. 3. The conclusion that we draw from Figs. 1, 2, 3 is that \(^4\text{He}\) forms a superglass.

Naturally, the results shown in Fig. 3 are influenced by a particular gas-like initial condition; another initial condition would produce a different result for \(\phi(\mathbf{r})\). Nonetheless, the fact that superfluidity and off-diagonal long-range order appear for just one such random initial condition, strongly suggests that these are genuine physical properties of the metastable phase.

Despite the above-mentioned fact that the MC dynamics is quite different from the real-time dynamics of helium (in simulations heat dissipates locally) it is possible to make semiquantitative arguments with regards to the stability of the superglass phase. Its stability on timescales several orders of magnitude longer than \(\omega_D^{-1} \sim 3^{-13}\) s, is guaranteed by observing no changes in the superglass properties over \(10^4\) MC sweeps. [The most appropriate physical interpretation of one sweep for the conventional PIMC scheme, is the time scale corresponding to the zero-point motion of atoms, whereby all particles have a chance to sample their optimal positions locally.] The extra advantage of using the worm algorithm is that pair-wise exchange, or tunneling of two particles, is sampled at the same rate as zero-point vibrations, while the rate \(J\) of exchange processes in the solid state of \(^4\text{He}\) is about five orders of magnitude slower (e.g., measured values of tunneling for \(^3\text{He}\) in \(^4\text{He}\) are of order \(J \sim 10 \mu\text{s}^{-1}[10]\)). It seems then plausible to assume that the metastability of the superglass phase extends up to \(10^4 J^{-1} \sim 1\) ms. If multiparticle tunneling (at low-temperature) events are required to reach the genuine equilibrium, then the actual degree of metastability has no obvious upper limit, and can easily exceed the experimental timescale.

Summarizing, we have provided theoretical evidence that \(^4\text{He}\) features a new metastable phase, a superfluid glass. This observation naturally suggests that other, more “regular” types of solid disorder, such as grain boundaries and dislocations, may also possess superfluid properties. We foresee further theoretical studies in the following directions:

1. Determination of the “phase diagram” of the glassy phase, i.e., of its domain of metastability (e.g., in the \((n, T)\) plane) and the line separating superglass from normal glass. The other important issue is to quantify the crossover line separating this novel, superglass phase from the superfluid: the superglass phase is characterized by the low-temperature plateau \(\rho_s(T) \rightarrow \rho_s < 1\), as in a “dirty” conventional superfluid.

2. Explore alternative possibilities of metastable supersolids: (i) A regular crystal doped with vacancies (or even interstitials). (ii) Superfluid grain boundaries and/or dislocations. (iii) The yet elusive superfluid phase of condensed para-hydrogen.

The superglass phase is not directly relevant to the interpretation of Kim and Chan’s experiment, since the MC temperature quench is much more rapid then in the experiment, leading to more disordered samples and
FIG. 3: Condensate wavefunction at $n=0.0359 \, \text{Å}^{-3}$ (represented by the density of points) and $T=0.2 \, \text{K}$, obtained by making ten slices of the system along the $\hat{z}$-direction and projecting them on the $xy$-plane (slices are ordered from left bottom to left top and then from right bottom to right top).

much larger superfluid fraction. However, a related scenario may be appropriate, namely that of a generalized superfluid-grain-boundary [7], which may include a foam-shaped superglass network interpenetrating the polycrystal.

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Note added. In an independent study [17], Clark and Ceperley calculated the single-particle density matrix of the ideal $\text{hcp} \, ^4\text{He}$ crystal at the melting density $n = 0.0287 \, \text{Å}^{-3}$. The data of Ref. [17] are consistent with ours for $n = 0.0292 \, \text{Å}^{-3}$.

[9] By definition, a solid is a state with broken translation symmetry. An immediate implication of broken translation symmetry is shear rigidity.
[15] In any case, small scale and the amount of disorder would make any such interpretation rather ambiguous.