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Supersolid State of Matter

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On the Supersolid State of Matter

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We prove that the necessary condition for a solid to be also a superfluid is to have zero-point vacancies, or interstitial atoms, or both, as an integral part of the ground state. As a consequence, superfluidity is not possible in commensurate solids which break continuous translation symmetry. We discuss recent experiment by Kim and Chan [Nature, **427**, 225 (2004)] in the context of this theorem, question its bulk supersolid interpretation, and offer an alternative explanation in terms of superfluid helium interfaces.

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Recent discovery by Kim and Chan [1, 2] that solid ^{4}He samples have a non-classical moment of inertia (NCMI) is a breakthrough result which has prompted renewed interest in the supersolid (SFS) state of matter. Early theoretical work by Andreev and Lifshitz [3] and Chester [4] showed that solids may feature a Bose-Einstein condensate of vacancies (or interstitial atoms) and thus possess superfluid (SF) properties. One may consider their work as establishing *sufficient* conditions for SFS. It was natural then to interpret mass decoupling in the torsion oscillator experiments as originating from small (about $\sim 1\%$) concentration of zero-point vacancies [1].

However, the overwhelming bulk of previous experimental work (for review, see, e.g., [5]) indicates that vacancies and interstitials in ^{4}He crystals are *activated* and their concentration is negligible below 0.2 K. The most recent study [6] looked at the density variations of solid ^{4}He between two capacitor plates and did not reveal any presence of vacancies. To deal with these facts an idea was put forward that strong exchange processes in quantum crystals may lead to superfluidity even in the absence of zero-point defects [7, 8]. Mistakenly, this idea is attributed to Leggett's paper [9]. Leggett established a link between the SF response and the *connectivity* of the groundstate wavefunction and derived a rigorous formula for the upper bound on the superfluid density, ρ_s. Crystal defects and their relation to the connectivity was *not* discussed in Ref. [9]. A separate issue is the "extremely tentative" order-of-magnitude estimate $\rho_s \leq 3 \times 10^{-4}$ based on the exchange integral between ^{3}He atoms [9], which, apparently, caused the misleading interpretation that interatomic exchange, on its own, may result in SF in the absence of vacancies [10].

The central point of the discussion to follow is to answer the question whether superfluidity is possible in crystal structures with the number of atoms being commensurate with the number of lattice points and what are the necessary conditions for this to happen.

Below we re-examine Leggett's work and show that it implies vacancies and/or interstitial atoms as a *necessary* condition for SFS in bosonic systems similar to ^{4}He. Chester's "final speculation" that without them the solid state of matter is insulating [4] proves to be a theorem. We present an alternative proof of the theorem using path-integral language in which the presence of vacancies in the SF state is seen explicitly. It also provides a simple picture showing why exchange processes on their own do not lead to SF. Thirdly, we put forward an argument based on the phase–particle-number uncertainty relation [11] which relates SF, compressibility of pinned solids and vacancies (pinning is crucial to separate and suppress the contribution to the compressibility coming from the change of the lattice constant from that due to adding/removing particles to/from the bulk [12]). The answer to the central question is then that SFS groundstates in commensurate solids have "zero measure" to be found in Nature, because they require an exact symmetry between zero-point vacancies and interstitials which is immediately broken by changing system parameters, e.g. pressure [13]. By excluding a bulk supersolid interpretation of the Kim & Chan results we are forced to look for an alternative explanation of their data based on the physics of disordered and frustrated ^{4}He interfaces.

As shown by Feynman [14], the groundstate of the bosonic system has no zeros, $\Psi_G(x_1, x_2, \ldots, x_N) \neq 0$. Moreover, in superfluids Ψ_G does not become macroscopically small when one or several coordinates, say, $x_1, x_2, \ldots x_m$, are taken around the system while other coordinates are kept fixed. This property (called connectivity by Leggett [9]) is key for superfluidity, and is just another way of saying that topological off-diagonal long-range order (TODLRO) is required for SF [15, 16, 17]. The requirement that connected Ψ_G be single-valued leads to the quantization of circulation and thus stability of persistent currents in samples with the cylindrical annulus geometry [9].

To illustrate the point, consider a one-dimensional system of two identical bosons forming a bound (molecular) groundstate, $\varphi_0(|r_1 - r_2|)$, with localization length l. Naively, the first rotating state of the molecule on a ring of large circumference, $L \gg l$, is written as a product of the the plane wave for the center of mass coordinate, $R = (r_1 + r_2)/2$, times the bound state: $\varphi_1 = e^{i2\pi R/L}\, \varphi_0(|r_1 - r_2|)$. This expression, however, is not single-valued, because if r_1 or r_2 is taken around the ring we get $\varphi_1 \to -\varphi_1$. The correct solution is to

replace φ_0 with $\tilde{\varphi}_0$, which has a zero at $|r_1 - r_2| \approx L/2$, i.e. in the region where φ_0 is exponentially small; at distances $|r_1 - r_2| \ll L$ the two functions are almost identical, $|\tilde{\varphi}_0| \approx \varphi_0$. Note, that the energetic cost of creating a zero in this case is exponentially small and vanishes in the limit of infinite system size.

The same considerations apply to the disconnected crystal state consisting of N bosons when Ψ_G decays to the macroscopically small value when any finite number of coordinates are taken away from their typical positions in the crystal (other coordinates are kept fixed) and moved around the system. The first rotating state in the system with periodic boundary conditions can be written as $\phi_1 = e^{i2\pi R/L}\,\tilde{\varphi}_0$ with $R = \sum_{i=1}^{N} r_i/N$ and $\tilde{\varphi}_0$ having zeros in regions where the modulus of φ_0 is exponentially suppressed and thus extra zeros do not cost finite (system size independent) energy. Clearly, the phase gradient circulation of ϕ_1 is $\sim 1/N$, and this system will not show the NCMI which is based on the impossibility of setting system in rotation with a macroscopically small velocity or circulation. This should be compared with the first rotating state of the single-atom superfluid system, $\phi_1^{(SF)} = e^{i2\pi \sum_{i=1}^{N} r_i/L}\,\varphi_0$ with $\varphi_0 > 0$ and phase gradient circulation 2π. Now, making zeros in connected φ_0 is so costly energetically that the lowest energy state corresponds to the relatively high kinetic energy of rotation. For definiteness, we consider below only single-atomic superfluids, but all considerations are readily generalized to the m-atomic case. We have essentially reproduced above the Leggett's argument that SF wavefunctions are necessarily connected.

By definition, $|\Psi_G(x_1, x_2, \ldots, x_N)|^2$ is the probability density to observe particles at the specified positions. We fix all coordinates except one, x_1, and observe that in connected wavefunctions $|\Psi_G(x_1)|^2$ remains finite when x_1 is taken arbitrary far from the initial position. Formally, this property is identical to statistical properties of atomic configurations in classical crystals at finite temperature and was used by Chester to introduce vacancies in the ground state. This final correspondence was not elaborated in Ref. [9].

How do we "visualize" vacancies/interstitials in the state of identical particles with strong exchange, *especially when the number of atoms coincides with the number of lattice points*? It appears that the common perception is that such solids do not have vacancies and interstitials, by definition, or else they are indistinguishable from the standard zero-point fluctuations. Imagine a solid sample pinned by an (arbitrarily weak) external potential preventing it from moving as a whole. There is no problem in identifying lattice points using the average particle density profile $\rho(r)$ which is periodic in space. Consider now some typical spatial configuration of particle positions along with the lattice points and start the coarse-graining procedure of "erasing" the closest particle-lattice point pairs in the spirit of the spatial renormalization group treatment. As we progress towards mesoscopic length-scales all short-ranged zero-point fluctuations of atoms away from lattice points will be erased from the picture. The procedure continues until we have erased all pairs with sizes much smaller than L but much larger than all microscopic scales. We say that the crystal state has no vacancies and interstitials if the final coarse-grained configuration is empty. On the other hand, if the coarse-grained configuration still contains lattice points, or particles, or both, at arbitrary large distances, we say that it has crystal defects in it. The decimation procedure explains how vacancies are possible in commensurate solids, and perfectly agrees with the conventional view of classical crystals at finite temperature. For the commensurate solid with connected Ψ_G we may start with the perfect-lattice configuration of particle coordinates and its empty coarse-grained picture, and then move x_1 arbitrary distance away to produce an image of the vacancy and interstitial. This will not result in the exponential suppression of the configuration probability (in fact, such configurations will dominate in the normalization integral).

In the absence of exact interstitial/vacancy symmetry the concentration of vacancies, n_v, and interstitials, $n_{\rm int}$, in the supersolid will be different, e.g. $n_v > n_{\rm int}$, since broken continuous symmetry allows production of excess vacancies by making small changes in the lattice constant. [This mechanism is is not available in discrete models, and then $n_v = n_{\rm int}$ is possible.] In the translation invariant system at T=0 one expects then $\rho_s = A n_v$, just like in any other interacting bosonic system.

Our second consideration is based on the path-integral formulation of quantum statistics [14] in terms of many-body trajectories, $\{x_i(\tau)\}$, in imaginary time $\tau \in [0, \beta]$ with periodic boundary conditions $\{x_i(\beta)\} = \{x_i(0)\}$. The most important superfluid characteristic of particle trajectories, or world lines, is their winding numbers, M^α, $\alpha = 1, 2, \ldots, d$. We assume periodic boundary conditions in space in all d-dimensions with linear sizes $L^\alpha = L$. To determine M^α imagine a cross-section going through point $\mathbf{R}$ perpendicular to the direction α and count how many times particles cross it from left to right, k_-^α, and from right to left, k_+^α. By definition, winding numbers are $M^\alpha = k_+^\alpha - k_-^\alpha$. They are independent of the cross-section location $\mathbf{R}$ because trajectories are continuous and periodic in imaginary time. The superfluid density is then given by [19]

$$\rho_s^{\alpha\gamma} = 2mTL^{2-d} \left\langle M^\alpha M^\gamma \right\rangle , \qquad (1)$$

where m is the particle mass. In $d = 3$, the superfluid density is finite in the thermodynamic limit, $L \to \infty$, $T \to 0$, and $T/L \to 0$, if the probability of having world lines with non-zero winding numbers in the ground state is close to unity.

We now demonstrate that crystal states without zero-point vacancies are described by world-line configurations with $\mathbf{M} = 0$, i.e. they are not superfluid. We start with the picture of a perfect crystal with particles tightly localized around equilibrium lattice points. In this state the trajectories are nearly straight lines with $\mathbf{M} = 0$ as

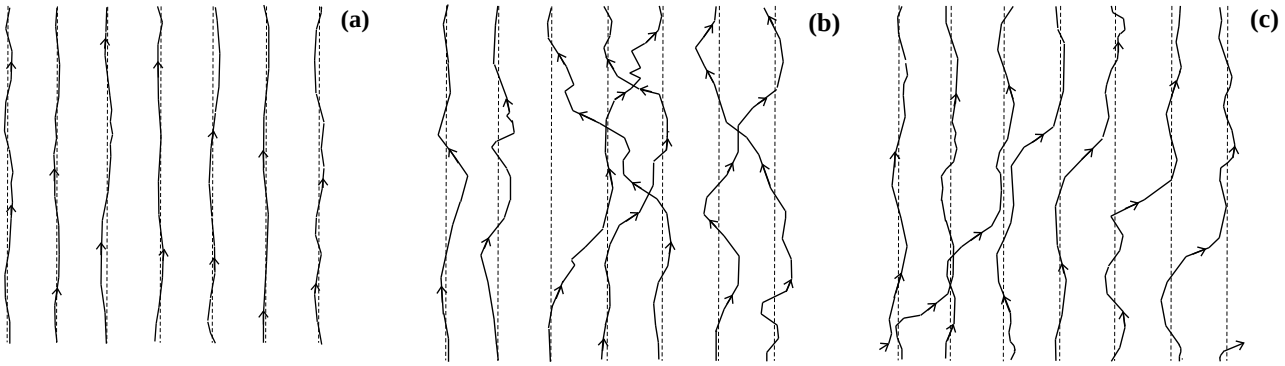

FIG. 1: Particle worldlines in different crystals at low temperature. The time axis is vertical. Dashed lines show the equilibrium lattice points. (a) Nearly classical crystal at low temperature; $\mathbf{M} = 0$. (b) Insulating quantum crystal with large zero-point fluctuations and frequent particle exchange processes; $\mathbf{M} = 0$. (c) Particle worldlines with non-zero winding number.

shown in Fig. 1(a). When tunnelling exchange processes are added into the picture the world lines are no longer in one-to-one correspondence with the lattice points. The nature of the exchange process, however, is such that when one particle leaves its equilibrium crystal point R_1 and goes to point R_2, the other particle goes from R_2 to R_1 (for pairwise exchange). Thus, the net current of world lines through any cross-section remains strictly zero. The same conclusion follows from the consideration of multiparticle exchange events [18], see Fig 1(b).

Consider now a world-line configuration with non-zero winding number, Fig. 1(c). At any moment of imaginary time we consider the spatial configuration of particle positions and lattice points and apply the coarse-graining procedure discussed above. Again, all short-lived and short-ranged exchange process and zero-point fluctuations will be erased once we pass several atomic distances, and in the insulating state the "movie" of the coarse-grained configuration evolution in time will show an empty "screen" for Figs. 1(a,b). If $\mathbf{M} \neq 0$, as in Fig. 1(c), there is no way for the renormalization procedure to erase all particles at all moments in time since topologically winding numbers correspond to particle trajectories moving continuously in the same spatial direction and thus creating imbalance between particles and lattice points at arbitrary large distances. After all other particles are associated with lattice points and erased, the winding trajectory describes an interstitial-vacancy pair which separates over the distance of order L and eventually makes a closed loop around the system, see Fig. 2. For the statistics of such loops to give non-vanishing $\langle M^\alpha M^\gamma \rangle$ in the thermodynamic limit, they have to be typical in a given lattice structure. In other words, zero-point vacancies and interstitials are an integral part of the groundstate.

Our last consideration is based on the relation between the *pinned* compressibility, κ, and zero-point vacancies. Compressibility can be calculated through the energies of states with one extra particle, E_{N+1}, and one extra vacancy, E_{N-1}, as $\kappa = 1/V\Delta E$, where V is the system

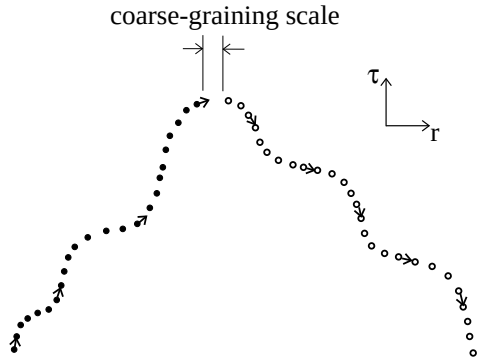

FIG. 2: Evolution of the coarse-grained picture for the trajectory with non-zero winding number similar to Fig. 1(c). Filled and open circles show particle and lattice site positions correspondingly. Arrows indicate the direction of the particle number current.

volume, and $\Delta E = E_{N+1} - E_{N-1}$. Incompressible states have finite ΔE. On another hand, in a macroscopic system ΔE can be obtained by considering the energy increase by creating an interstitial-vacancy pair with arbitrary large separation between them (in pinned solids the notion of vacancy or interstitial is rigorously defined within the coarse-graining procedure as the presence or absence of perfect registry between particles and unit cells). Crystals without zero-point defects are gapped with respect to the interstitial-vacancy production (otherwise these defects would be an essential part of the groundstate) and thus are incompressible (if pinned) and *vice versa*. One step further, this implies that superfluidity and pinned compressibility come together and either one (including long-wave acoustic properties with additional sound mode) can be used for the detection of the SFS state experimentally. This conclusion is in line with the famous uncertainty relation [11] between the phase of the superfluid order parameter, ϕ, and particle number, $\Delta\phi\Delta N \geq 1/2$. Because of this relation, one may not

introduce a well defined phase field for the incompressible state of matter which tends to *completely* suppress particle number fluctuations.

We have little doubt that large activation energies for vacancies and interstitials in ^{4}He measured down to $\sim 1\ K$ temperatures [5] will not radically change to near zero at lower temperatures, and that helium is a commensurate solid at $T = 0$. Since it has no symmetry between the vacancies and interstitials, we conclude that there are no zero-point vacancies in bulk solid ^{4}He. By excluding superflow through the crystal bulk we are forced to look more closely at the superfluid properties of disordered helium-substrate layers and frustrated interfaces between helium micro-crystallites.

There are indications of strong disorder in experimental system of Ref. [1]. The dependence of the superfluid density on reduced temperature parameter $t = (T_c - T)/T_c$ has little to do with the expected bulk superfluidity $t^{0.671}$ dependence. Instead, ρ_s appears to vanish at T_c with zero derivative. Such a behavior can be modelled by a broad distribution of transition temperatures in the heterogeneous sample. This observation correlates with the gradual decrease of the decoupled mass with the increase of the torsion oscillator amplitude by orders of magnitude. Let us assume that a sample consists of micro-crystallites of linear size D with superfluid interfaces of typical thickness d between them. The superlfuid fraction may be estimated then as $\rho_s/\rho \sim d/D$. To have $\sim 1\%$ of the superfluid mass coming from interfaces with $d \sim 10\ \mathring{A}$ one will need crystallite sizes about a fraction of a micron. The variety of interfaces with different crystallographic indices provides a broad distribution of transition temperatures.

One of the experimental mysteries is extreme sensitivity to the addition of ^{3}He impurities at the level of $n_{\rm im} \sim 100 ppm$. To minimize kinetic energy, light ^{3}He atoms are likely to end up at frustrated interfaces, and then at the edges where different interfaces meet. This may increase ^{3}He edge *vs* bulk concentration by a factor as large as $(D/d)^2$ and produce $n_{\rm im}^{\rm (edge)} \sim 1$. This will have a profound effect on the edge-connected interface superfluidity (edges then act as a disordered two-dimensional network of Josephson junctions).

We are not aware of any systematic study what are the properties of interfaces between the ^{4}He micro-crystals and helium ^{4}He crystals on disordered substrates at elevated pressures. Model simulations of domain wall boundaries in the checkerboard solid (obtained for interacting lattice bosons at half-integer filling factor) show that they remain superfluid deep into the bulk solid phase [20]. In the outlined picture three predictions are certain: (i) the superfluid fraction must strongly depend on the crystal growth process, (ii) the amount of ^{3}He sufficient to suppress superfluidity scales as $n_{\rm im} \propto \rho_s^2$, and (iii) transition temperatures on interfaces do not depend on D and $\rho_s(T = 0)$.

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