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Bose-Hubbard model and superfluid staircases in ^{4}He films

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The recent experimental observation of steplike structure in the superfluid density of ^{4}He films on graphite is interpreted in terms of passage close to Mott insulating lobes in the Bose-Hubbard model. Plateaus develop in the superfluid density near the completion of each layer because of the He-He repulsion. We modify the Bose-Hubbard Hamiltonian, using a density-dependent He-He interaction term to model the He-substrate potential. Quantum Monte Carlo simulations yield results that agree well with the experimental data.

^{4}He films have been valuable in studying quantum-mechanical effects in systems of reduced dimensionality.[1] In particular, there has been a large body of work on superfluidity in thin films.[2] Most of these experiments have been conducted on disordered substrates. A recent paper,[3] however, reported on the measurements on the superfluid density in ^{4}He films adsorbed on the basal plane of graphite, which is ordered on atomic length scales. A number of features were observed, including the appearance of superfluidity in a narrow range of coverage in the second layer, and a subsequent staircase structure in the superfluid signal as a function of coverage. The plateaus in this staircase appear near completion of the third to sixth layers. This discovery complemented the vapor pressure and third sound measurements of Zimmerli, Misture, and Chan,[4] which showed that the chemical potential μ also grows in steps out to the completion of the seventh layer and that the third sound velocity has structure that is periodic in coverage. The steps in μ occur at layer completion, where the superfluid signal as measured in the torsional oscillator experiment[3] is nearly constant.

Meanwhile, a body of analytic and numerical work[5–13] on models of interacting bosons, with and without disorder, has established the corresponding phase diagrams and the critical properties of these systems. The "Bose-Hubbard" (BH) Hamiltonian

$$H=-t\sum_{\langle ij\rangle}(a_i^\dagger a_j+a_j^\dagger a_i)-\mu\sum_i n_i+V\sum_i n_i^2 \qquad (1)$$

is believed to represent much of the qualitative physics of the helium system, and upon incorporating disorder, for the related problem of granular superconductors.[5] Here a_i is a boson destruction operator at site i, and $n_i=a_i^\dagger a_i$. The transfer integral t sets the scale of the kinetic energy, while μ and V are the chemical potential and the boson-boson repulsion.

In this paper we suggest analogies between the dependence of $\mu(n)$ and the superfluid density $\rho_s(n)$ on the density n in ^{4}He films adsorbed on graphite and in the BH model. It has been established previously that the BH model exhibits a steplike structure in the chemical potential as a function of n.[5] To understand this, consider the empty lattice for the case $t=0$. Particles can be added to the system at no cost, i.e., $\mu=0$, until each site is occupied by a single atom. Once each site is occupied, however, additional atoms must be given an energy V to overcome the repulsion of the atom which is already present. Continuing this argument, we see that the chemical potential develops a jump, which is proportional to V, at the commensurate densities $n=1,2,3,\ldots$ atoms/site and is constant for all intermediate densities. These jumps in μ also imply the appearance of a gap Δ in the spectrum. For the case $t>0$, the steps decrease in size, and the chemical potential is no longer constant at incommensurate fillings.

The presence of the gap for sufficiently small t/V, i.e., large interaction strengths, means that the interparticle repulsion V *localizes* the bosons into Mott-type insulating phases at commensurate fillings. Inside these phases the superfluid density ρ_s vanishes. With increasing t/V, the benefit in exchange energy eventually overcomes the repulsive cost of delocalizing the bosons at some critical value t_c. For $t>t_c$ the bosons become extended and form a superfluid at low temperatures. At incommensurate fillings, the bosons are delocalized at all hopping strengths and will always be superfluid. The qualitative phase diagram for clean systems is shown in Fig. 1.[5] This phase diagram has recently been confirmed quantitatively by Monte Carlo simulations,[7,8] by coarse graining methods,[12] and by low-order perturbation expansions.[13]

The staircase structure in the superfluid density ρ_s can also be understood in the context of the BH model. As discussed above, the superfluid density vanishes inside and at

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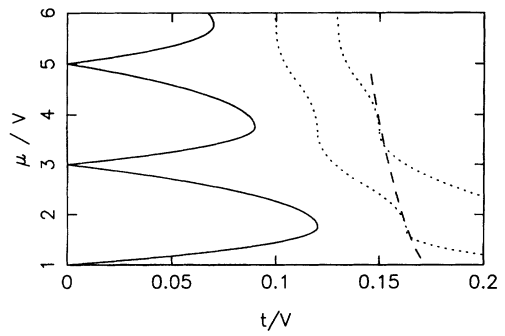

FIG. 1. Qualitative phase diagram of the Bose-Hubbard model. The solid lines are the boundaries of the localized Mott lobes. The dotted curves are contours of constant superfluid density. The approximate trajectory followed in our simulations, in which V depends on density in a way described in the text, is shown as a dashed curve.

the boundaries of the Mott insulating lobes in Fig. 1. The superfluid density is expected to grow smoothly with μ when the system is deep in the superfluid part of the phase diagram. In Fig. 1 we show qualitatively the contours of constant ρ_s using dotted curves.[14] As the density is increased, the system follows a nearly vertical path in parameter space similar to that shown by the dashed curve in Fig. 1. The choice of this path will be discussed below. If this trajectory passes in the vicinity of the Mott lobes, it will be nearly tangential to the contours, resulting in plateaus in ρ_s vs n.

The BH model is only a very simplified approximation of a real ^{4}He film. Nevertheless, the model captures the correlation effects near the insulating lobes which cause the staircase structure. If we measure ρ_s as a function of ρ at constant V (a vertical sweep in Fig. 1), well defined plateaus are observed. However, these plateaus get weaker somewhat too rapidly as the coverage increases, as compared to the experiments. There are several ways to refine the model to address this. First, the BH model does not incorporate the long-range attractive tail of the He-He potential. As a consequence, none of the *structural* transitions[15,16] that occur in the first two layers of adsorbed ^{4}He appear in the BH phase diagram. As we are interested in superfluid properties, and the first two layers are known to solidify,[16,17] we will only study the system at coverages above two layers. We consider the first two layers to form a pseudosubstrate for the overlying superfluid film, and we use the BH Hamiltonian only to model layers added above the two solidified layers. Therefore the first layer in our calculations will correspond to the third experimental layer. A second shortcoming of the BH model, which we address here, is the exclusion of the He-substrate potential. Two common approaches to the He-substrate interaction are the Dzyaloshinskii-Lifshitz-Pitaevskii (DLP)[18] and Frenkel-Halsey-Hill (FHH)[19] theories. In their original formulation, both of these theories model the fluid and the substrate as structureless continua, disregarding the effects of layering as well as structural phase transitions in the film. They consider the shift in the chemical potential, $\Delta\mu$, caused by the presence of the smooth surface. In both cases the result can be cast in the form:

$$\Delta\mu(d) = -\gamma(d)/d^3, \tag{2}$$

where the coefficient $\gamma(d)$ is a function of the film thickness d. This form can be derived by assuming a van der Waals interaction between the helium and the surface. The FHH theory adopts the simplest assumption: $\gamma(d)=\gamma(0)$, a constant.

DLP provide a generally formulated many-body approach. As shown by Cheng and Cole,[20] the FHH formula is the first term upon expanding the DLP theory in the limit of small densities and polarizability, neglecting retardation. The higher-order terms of the DLP expansion can be neglected at the thicknesses under consideration here.[20] On the other hand, Zimmerli, Mistura, and Chan have shown that the FHH formula overestimates the film thickness by nearly a full layer.[4] A reasonable modification has been proposed by Cheng and Cole,[21] who treat the solid layers as a pseudosubstrate, leading to a modified $\gamma(d)$:

$$\gamma(d') = 1772K\text{Å}^3\{0.07+[1+D/(d')]^{-3}\}, \tag{3}$$

where $D=4.42$ Å is the thickness of the first two solid layers and $d'=d-D$ is the thickness of the fluid film. The parameters used in Eq. (3) were extracted by Cheng and Cole from experiments.[21] Measurements of the third sound velocity by Zimmerli, Mistura, and Chan[4] provided quantitative support for this picture. We will use Eqs. (2) and (3) to represent the effects of the substrate and the two solid layers on the chemical potential of films thicker than two atomic layers.

While it may be possible to introduce layers in the DLP formalism, the Bose-Hubbard model provides a natural framework to study such layering effects, and therefore we concentrate on the BH model in this paper. We solved for the equilibrium thermodynamic properties of the BH Hamiltonian using the Quantum Monte Carlo (QMC)[22] technique. This approach evaluates operator expectation values, treating the many-body correlations in an exact manner by computing the imaginary time evolution operator $e^{-\beta H}$ using stochastic techniques. The results presented here are for 8×8 spatial lattices with a discretization of imaginary time equal to 1/32 of the bandwidth. Convergence of the results and finite-size effects have been carefully checked.

Representing the layers individually and introducing the substrate-^{4}He potential would constitute a full treatment of the problem. It would, however, be costly computationally, limiting the lattice size and raising the minimum temperature that could be studied. As our main emphasis is on the many-body and commensuration effects, which are most prominent for large systems and low temperatures, we chose instead to model the density dependence of the chemical potential by a density-dependent interaction term $V(n)$, where $n=\langle n_i\rangle$ is the density in ^{4}He atoms/site. The underlying idea is that the interaction term in the Hartree approximation, $H_{\text{int}}=\Sigma_i V(n)n_i^2 \approx \Sigma_i V(n) n n_i$, can be thought of as renormalizing μ. We use the relation

$$V(n) = V+\Delta\mu[d'(n)]/n = V-C\left(\frac{1}{n^4}+\frac{0.07}{n(n-2)^3}\right), \tag{4}$$

valid for densities greater than 2 atoms/site. We regard both V and C as phenomenological parameters, to be determined below. We emphasize that when the full chemical poten-

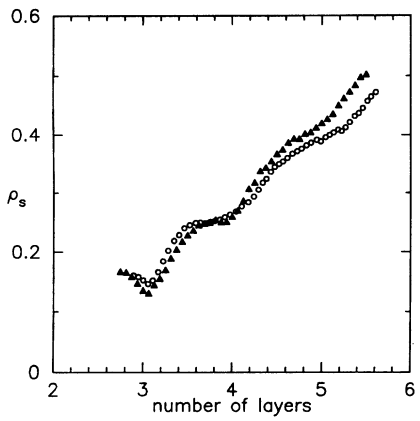

FIG. 2. The superfluid density ρ_s at 500 mK determined from torsional oscillator measurements (circles) and the quantum Monte Carlo simulation (triangles). The experimental data has been scaled by a constant and its coverage scale has been shifted as described in the text.

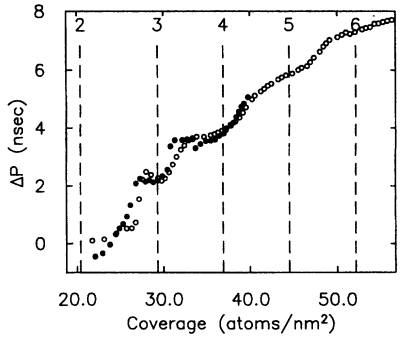

FIG. 3. The period shift data at 20 mK (closed circles) and 500 mK (open circles) are shown as a function of the ^{4}He coverage. The dashed lines indicate layer completion according to the coverage scale used in Fig. 2

tial is extracted from the ground-state energy E_G, via $\mu = \partial E_G / \partial n$, this smooth shift will be further renormalized by the effects of the He-He interaction, leading to the staircase structure.

We compare the results of our calculation with the torsional oscillator measurements of Crowell and Reppy.[3,23] Their experimental apparatus comprises a cell containing exfoliated graphite mounted on a torsion rod. The measurements discussed here were made while adding ^{4}He to the cell at constant temperature. For each coverage, the superfluid period shift ΔP was determined by taking the absolute value of the difference between the resonant period and the period that would have been measured if the film were non-superfluid and hence locked to the substrate. The period shift ΔP is proportional to the superfluid density ρ_s if the film is a uniform fluid. The experimental technique is discussed in greater detail elsewhere.[24]

In Fig. 2, we show the results of a sweep for $V/t = 8.5$ and $C/t = 88$ compared with the experimental period shift data at 500 mK, which have been scaled by a constant. This figure displays the central result of this paper, namely, the close reproduction of the plateau structure of ρ_s in the experiments by quantum simulations of the Bose-Hubbard model. Although the best fit of the data to the theory was obtained for the above values of V/t and C/t, the plateau structure appears over a fairly wide range of these parameters. The plateaus are less sensitive to the choice of C, appearing even in the case $C = 0$, for which $V(n) = V$. Our value for β corresponds to a T on the order of the bulk transition temperature.[25]

The agreement of the calculation with the experimental data suggests that the origin of the plateaus in ρ_s is the localizing effect of the interparticle repulsion close to the completion of each layer. The suppression of the mobility of the bosons in the superfluid phase reflects the proximity of the insulating Mott lobes. Although the overall plateau structure is a robust feature of the model, the dip in the numerical calculation near $n = 3$ is due to the close proximity of the trajectory to the Mott lobe and is therefore sensitive to the choice of parameters. We emphasize that ρ_s is not simply proportional to the areal density of ^{4}He. With the exception of the plateau at third-layer completion, the plateaus start before the completion of each layer, even though the density of the layer continues to increase.

Additional evidence supporting our conclusions can be found in the experimental data at 20 mK, which are shown along with the 500-mK data in Fig. 3. The plateau at $n = 4$ is even more pronounced with decreasing T. This observation is consistent with the fact that the localizing effects of the repulsion are enhanced at lower temperatures.[6,12] We did not carry out numerical simulations at 20 mK because of the prohibitively long computation time.

The plateau at third-layer completion may have a different origin, a structural phase transition in the film. Lauter *et al.*,[17] using neutron scattering, have observed a sudden increase in the density of the second-layer incommensurate solid in the vicinity of third-layer completion. The atoms that go into the second-layer solid in this case come at the expense of the fourth layer, which does not start to fill until the reconstruction of the second layer is complete. The experimental data are consistent with this hypothesis, since there is no indication that the atoms added to the film between 28 and 30 atoms/nm^2 make a contribution to the superfluid signal. The plateau near third-layer completion is also much narrower than those found at higher coverages and its onset occurs closer to layer completion, suggesting that the underlying mechanism may be different from that responsible for the behavior observed at higher coverages. If the reconstruction occurs, the coverage at third-layer completion should be shifted upward from the conventional value of 28.0 atoms/nm^2. We have done so in Fig. 2, finding the best agreement between theory and experiment when the fourth layer starts filling at 29.3 atoms/nm^2. This shift is somewhat greater than the lower bound of 0.8 atoms/nm^2 inferred from the neutron scattering data of Lauter *et al.*,[17] but it is less than $\sim 30\%$ of the width of the plateau at fourth-layer completion.

An alternative scenario for the plateaus has been suggested by Clements *et al.*[26] They find that liquid clusters form at low densities in the third, fourth, and fifth layers. These do not contribute to the superfluid signal until they percolate across a macroscopic region of the substrate. As a result there is a plateau in ρ_s at the beginning of each layer,

followed by a rapid increase once percolation occurs. The clustering theory predicts the appearance of plateaus *after* layer completion. However, the plateaus in the experimental data corresponding to the fourth, fifth, and sixth layers start several atoms/nm^2 *before* layer completion. We emphasize that even without any reconstruction shift, our numerical simulations yields plateaus which begin substantially before layer completion, in agreement with the experiment.

In summary, we have considered recent experiments showing the formation of plateaus in the superfluid density of thin films of ^{4}He on graphite. Although other mechanisms may be important at lower coverages, we find that the formation of plateaus near the completion of the fourth and higher layers is driven primarily by the localizing effect of the strong He-He repulsion. Our simulations, which incorporate the He-substrate potential in an approximate way, agree quantitatively with the experimental data. Improvements on the model, such as accounting for the long-range attractive part of the He-He interaction, may enhance the agreement with the data at lower densities.

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