Phase Separation in Supersolids

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We study quantum phase transitions in the ground state of the two dimensional hard-core boson Hubbard Hamiltonian. Recent work on this and related models has suggested "supersolid" phases with simultaneous diagonal and off-diagonal long range order. We show numerically that, contrary to the generally held belief, the most commonly discussed "checkerboard" supersolid is thermodynamically unstable. Furthermore, this supersolid cannot be stabilized by next-near-neighbor interaction. We obtain the correct phase diagram using the Maxwell construction. We demonstrate that the "striped" supersolid is thermodynamically stable and is separated from the superfluid phase by a continuous phase transition.

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Some 40 years ago, Penrose and Onsager [1] posed the question: Is it possible for a bosonic system, like ^{4}He, to have a phase where long range crystal order ("solid") and off-diagonal long range order (superfluidity) coexist? Their answer, based on an analysis which ignored zero-point fluctuations, was that such supersolid phases do not occur. Subsequently, it was argued [2–4] that including the effect of large zero-point quantum fluctuations in the crystal phase might allow for the existence of a supersolid. This question has continued to spark much theoretical, numerical, and experimental interest [5]. There has been a convergence of agreement, based on mean-field [3,6–8] and numerical [8–12] work, that supersolids do exist, in 2D lattice models, particularly in systems in which the density of bosons is doped away from the commensurate fillings which are optimal for charge ordering. The existence of supersolids for 2D quantum bosons has fundamental implications to vortex phases in superconductors because of formal mappings between the problems [13]. In this paper, we demonstrate that the most discussed of these lattice supersolid phases is thermodynamically unstable, and we argue that it does not exist in any region of interaction strength or density.

Consider a 2D square lattice with one hard-core boson for every two sites ($\rho = \frac{1}{2}$) interacting with near-neighbor (nn) repulsion. If the interactions are weak, the bosons will be mobile and condense into a superfluid phase at low temperature. If the repulsion is strong, the system will freeze into a charge density wave pattern in which sites are alternately occupied and empty. At $\rho = \frac{1}{2}$ these possibilities are mutually exclusive. If we remove or add a boson, the resulting bosonic defect could "hop" among the background of charge ordered particles if the zero-point fluctuations are large enough. A dilute gas of such defects may Bose condense and form a superfluid superimposed on the background of crystal order, a "supersolid" phase. If, instead, next-near-neighbor (nnn) repulsion dominates, the charge ordering is in stripes, but the basic issue of a condensation of additional bosons coexisting with a striped pattern is as for checkerboard. While it is useful to think of separate frozen and superfluid bosons, these quantum particles are indistinguishable. All the bosons simultaneously participate in both types of long range order.

Calculations supporting this intuitive physical picture are primarily based on mean-field theory with spin wave stability analysis. They initially dealt with checkerboard charge order where the ordering vector for the structure factor is $\mathbf{k} = (\pi, \pi)$. Liu and Fisher [6] argued that the supersolid phase exists for hard-core bosons with nn repulsion, but that it is unstable in the sense that the *critical velocity* vanishes. If nnn repulsion is present, the supersolid can be stabilized [14]. Numerical and analytical studies of the quantum phase model (QPM) [15], which describes soft-core bosons, showed that the (π, π) supersolid phase exists even without nnn repulsion, due to the soft cores. The supersolid is present even at half filling, i.e., in the absence of any defects [10]. Simulations of the *hard-core* bosonic Hubbard model similarly found that the supersolid phase exists in the absence of nnn repulsion off half filling, but unlike the QPM is absent at half filling. In addition, a mean field with spin wave analysis showed that this supersolid phase has a finite critical velocity [8,11]. However, it is generally accepted that at least in the presence of nnn repulsion, the (π, π) supersolid phase is stable.

Discussions of stability based on nonvanishing critical velocity examine the effects of low energy excitations on an *existing* supersolid phase. To our knowledge, however, there has been no discussion or numerical verification of the underlying thermodynamic stability of either of these supersolid phases against phase separation. Simulations done at *fixed* particle number which found simultaneous diagonal and off-diagonal long range order, [8,10,11,16,17], do not address the possibility of phase separation. In what follows we examine thermodynamic stability of the checkerboard and striped supersolids by constructing the chemical potential particle number relation and calculating the compressibility.

We use a new dual quantum Monte Carlo algorithm [18] to simulate the hard-core bosonic Hubbard model,

$$H = -t\sum_{\langle ij\rangle}(a_i^\dagger a_j + a_j^\dagger a_i) + V_1\sum_{\langle ij\rangle}\hat{n}_i\hat{n}_j + V_2\sum_{\langle\langle ik\rangle\rangle}\hat{n}_i\hat{n}_k\,. \quad (1)$$

a_i ($a_i^\dagger$) are destruction (creation) operators of hard-core bosons on site i of a 2D square lattice, and n_i is the density at site i. The hopping parameter is chosen to be $t = 1$ to fix the energy scale. V_1 (V_2) is the near-neighbor (next-near-neighbor) interaction. At $V_2 = 0$, and after an appropriate sublattice spin rotation, this boson model is equivalent to the spin-$\frac{1}{2}$ antiferromagnetic XXZ model. In this language, superfluid order corresponds to magnetic order in the XY plane, while density order corresponds to magnetic order in the Z direction.

To determine numerically the nature of the ground state of (1), we evaluate, at fixed particle density, the equal time structure factor at the ordering vector $\mathbf{q}$,

$$S(\mathbf{q}) = \frac{1}{N}\sum_{\mathbf{l}} e^{i\mathbf{q}\cdot\mathbf{l}}\langle n(\mathbf{j},\tau)n(\mathbf{j}+\mathbf{l},\tau)\rangle\,, \quad (2)$$

and the superfluid density [11,19], $\rho_s = \langle W^2\rangle/2\beta$, where W is winding number of the boson world lines. Ground state results for $S(\mathbf{q})$ and ρ_s are shown in Fig. 1 (Fig. 2) for the checkerboard (striped) phase. In both cases ρ_s is nonzero everywhere except precisely at half filling, but $S(\mathbf{q})$ also remains large off half filling, indicating solid order. Using finite size scaling to extrapolate to the limit of infinite lattice size for these fixed density systems [8,10,11,16,17], one can show that density correlations are indeed still long ranged off half filling where $\rho_s \neq 0$. The conclusion is that checkerboard and striped supersolid phases exist in the thermodynamic limit.

Already, however, it was remarked [8] that the energy versus density curves had small negative curvature in the (π,π) supersolid phase. It was speculated that this was evidence for phase separation. The $(\pi,0)$ supersolid phase showed no such negative curvature. It was recently shown numerically [17] that the easy-axis spin-$1/2$ XXZ model on a square lattice exhibits a first order spin-flop transition. These results confirm the discontinuous nature of the transition from the superfluid phase as ρ is adjusted in the *absence* of nnn repulsion.

To address the possibility of phase separation systematically, i.e., to obtain the phase diagram in the interaction-chemical potential (μ) plane, we must obtain ρ as a function of μ and use the Maxwell construction [20]. We first study the checkerboard case by fixing $V_2 = 0$ and scanning the filling for several values of V_1. The chemical potential for n bosons is calculated from the total energy: $\mu(n) = E(n+1) - E(n)$. We work on lattices up to size 12×12, and at temperatures as low as $\beta = 6$ to access the ground state properties.

Figure 3 shows ρ versus μ for $V_1 = 3, V_2 = 0$, as in Fig. 1. The slope of this curve is the compressibility, $\kappa = \partial\rho/\partial\mu$. Two $\kappa < 0$ branches are clearly seen just before and after the energy gap. The gap itself corresponds to the incompressible (π,π) solid at half filling which is seen in Fig. 1. Using the Maxwell construction we find the critical value of the chemical potential, μ_c (vertical dashed line), and read off the critical filling ρ_c in Fig. 3. The structure factor (Fig. 1) begins a very rapid rise at the point where κ turns negative [21]. It is crucial to note that ρ_s and $S(\pi,\pi)$ are both nonzero (Fig. 1) *only* in the *unstable* $\kappa < 0$ region

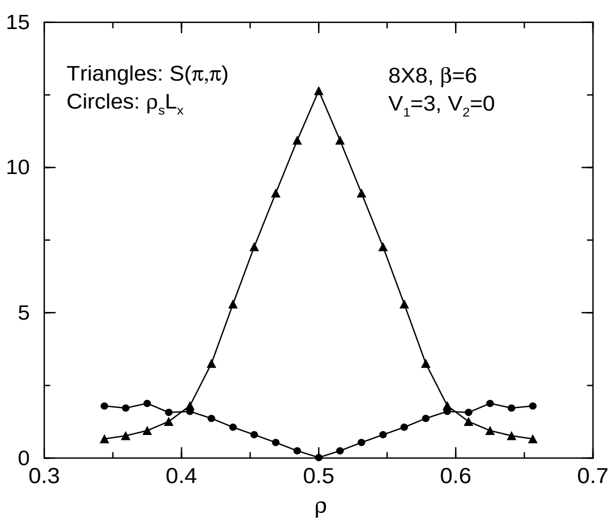

FIG. 1. The structure factor $S(\pi,\pi)$ and ρ_s as a function of fixed density. The half-filled point $\rho = 0.5$ is a solid with $\rho_s = 0$. For ρ close to $\rho = 0.5$, $S(\pi,\pi)$ and ρ_s are both nonzero. Moving away from half filling, eventually $S(\pi,\pi)$ will no longer scale linearly with system size and the system is a pure superfluid.

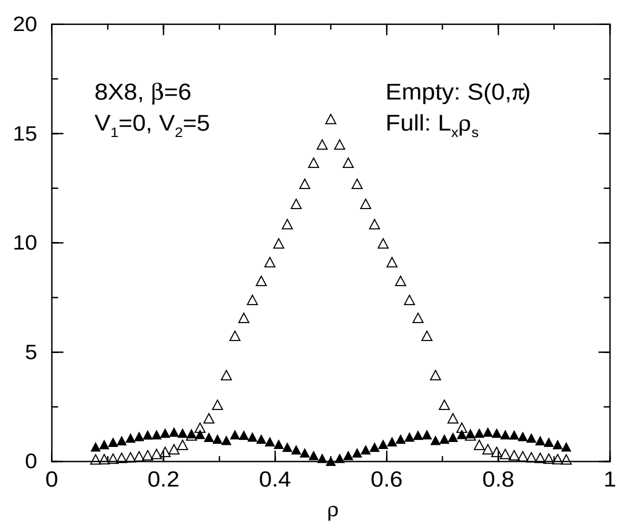

FIG. 2. The structure factor $S(0,\pi)$ and ρ_s as a function of fixed density. The half-filled point $\rho = 0.5$ is a solid with $\rho_s = 0$. For ρ close to $\rho = 0.5$, $S(0,\pi)$ and ρ_s are both nonzero. As with the checkerboard case, finite size scaling for $S(0,\pi)$ determines the density at which solid order vanishes.

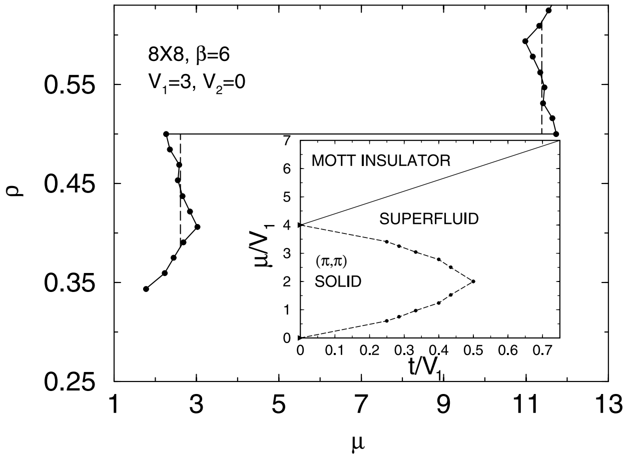

FIG. 3. ρ versus μ showing the $\kappa < 0$ regions. The vertical dashed line shows the location of the transitions from the Maxwell construction. Inset: The phase diagram for $V_2 = 0$. The solid line shows the continuous transition to the Mott phase at full filling; the dashed line shows the discontinuous transitions from the superfluid to the checkerboard solid at half filling. The density changes discontinuously across this line. The tip of the lobe is a continuous critical point.

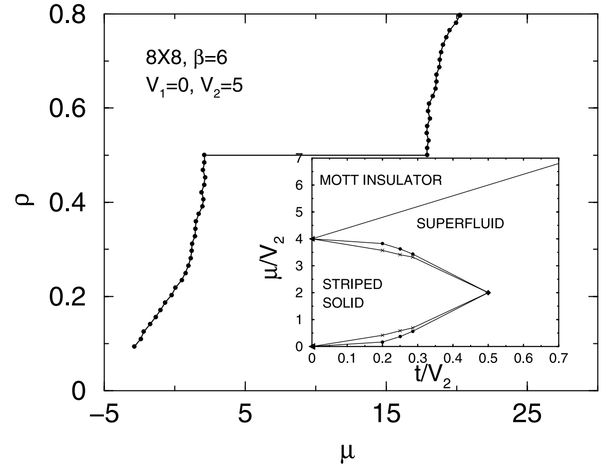

FIG. 4. ρ versus μ. Inset: The phase diagram for $V_1 = 0$. The narrow regions sandwiched between superfluid and $(\pi, 0)$ solid phases are the stable supersolid phases.

(Fig. 3). On one side of the transition the phase is purely superfluid while on the other side it is a (π, π) solid. What was previously accepted to be the checkerboard supersolid at fixed density, lies entirely on the $\kappa < 0$ branches and therefore phase separates into a mixture of solid and superfluid phases at densities $\rho = \rho_c$ and $\rho = \frac{1}{2}$. The metastable states in Fig. 3 correspond to either the superfluid or the gapped insulating phases.

To check if nnn repulsion stabilizes this phase against phase separation, we did simulations [21] with $V_1 = 3$ and V_2 ranging from deep in the (π, π) solid region to close to the boundary with striped order. We found the same $\kappa < 0$ behavior as in Fig. 3. Next-near-neighbor repulsion *does not* stabilize the checkerboard supersolid phase.

Repeating the simulations that gave Fig. 3 for different values of V_1 we construct the phase diagram in the $(\mu/V_1, t/V_1, V_2 = 0)$ plane, shown as the inset in Fig. 3. As the tip of the lobe is approached, the energy gap opens without $\kappa < 0$ regions in the ρ, μ plane. Therefore this point is apparently a continuous transition. This is consistent with Ref. [22] while Ref. [23] finds a first order transition in a model with longer range (Coulomb) interactions.

The same analysis for $V_1 = 0$, scanning V_2 and ρ, determines the stability of the striped supersolid. Figure 4 is a typical plot of ρ versus μ traversing the incompressible (gapped) striped solid at $\rho = \frac{1}{2}$. This is strikingly different from Fig. 3. There is no $\kappa < 0$ region: The phase transitions are all continuous. Furthermore, as μ is increased from the lowest shown value, the slope, i.e., κ, changes markedly at $(\mu \approx 0.74, \rho \approx 0.25)$. We find that $S(0, \pi)$ (Fig. 2) begins a rapid increase, indicating long range striped order, at precisely this particle density, ρ. Since the superfluid density, ρ_s, is still finite, we conclude that the rapid crossover in κ, like the behavior of the structure factor, signals the continuous transition from the superfluid to the $(0, \pi)$ supersolid. Increasing μ further takes the system into the gapped $(0, \pi)$ insulator, then back to the striped supersolid, and finally to the superfluid phase at $\mu \approx 19.2$. At strong coupling, the gap (the jump in μ) across the incompressible striped phase is $4V_2 = 20$ for the 2D square lattice. This value is reduced by quantum fluctuations as V_2 decreases, eventually disappearing entirely at weak coupling. The absence of negative compressibility regions indicates that all phases, in particular, the striped supersolid, are thermodynamically stable.

Repeating these simulations for various values of V_2 gives us the phase diagram in the $(\mu/V_2, t/V_2, V_1 = 0)$ plane (inset in Fig. 4). The regions sandwiched between the superfluid and the striped solid are the two stable striped supersolid phases. As the tip is approached, the supersolid phase gets narrower since the supersolid-$(0, \pi)$-solid transition approaches the supersolid-superfluid (SS-SF) transition. This prevents us from resolving these transition points near the tip. The difficulty in the numerical determination of these points stems from the fact that when the SF-SS and SS−striped solid transitions get very close to each other, they start behaving numerically as multicritical points. However, it appears that the supersolid phase completely surrounds the striped solid phase except at the tip and the base where we have multicritical points.

We also studied the effect of nn repulsion on the $(0, \pi)$ phase and found that the $(0, \pi)$ supersolid remains stable and that additional gapped phases appear at other special fillings [10,24]. Whether there are associated supersolid phases is under investigation [24].

The fact that it is easier to support nonzero ρ_s in a striped solid than a checkerboard one can be qualitatively argued as follows: In a striped solid doped away from half filling, defects have channels in which they can move at no interaction energy cost, and, importantly, the kinetic energy of these defects is set by t and can be controlled independently of the strength of the interaction V_2 which determines the solid order. In a checkerboard solid, the motion of a defect proceeds through an intermediate state of energy $2V_1$, giving a reduced effective hopping $t_{\mathrm{eff}} \approx t^2/2V_1$. V_1 controls simultaneously the defect kinetic energy and the tendency to charge order. As a consequence, there is reduced ability to tune to a supersolid phase. It is still remarkable, though, that the striped phase forms even at very low densities. Indeed, in the fermion Hubbard model, very small doping (just a few percent) away from half filling destroys long range spin order (antiferromagnetism), leaving little possibility that it might coexist with superconductivity.

In this paper, we have presented quantum Monte Carlo results for ground state correlations in the hard-core bosonic Hubbard model with near- and next-near-neighbor repulsion. We show that the $\mathbf{q} = (\pi, \pi)$ checkerboard supersolid, contrary to current beliefs, is an unstable phase and does not exist thermodynamically for this model for any filling or nnn repulsion. Instead, the system phase separates into solid and superfluid phases. This contradicts mean-field predictions which examine stability via an evaluation of the critical velocity for spin waves, and we find that in the presence of next-near-neighbor repulsion the supersolid is stable. We have not examined the soft-core case in detail, but preliminary results indicate negative compressibility regions in that case, too [8]. The quantum phase model for soft-core bosons also exhibits negative compressibility phases [25].

We found the striped supersolid phase $\mathbf{q} = (\pi, 0)$ to be stable and separated from the superfluid phase by a continuous transition. The energy $E(n)$ provides a signal of the transition: The compressibility exhibits a rapid crossover from the superfluid to the supersolid phase, with $\kappa_{\mathrm{SF}} < \kappa_{\mathrm{SS}}$. The issue of the stability of possible supersolid phases at other densities and wave vectors which are associated with the presence of long range interactions is a fascinating one which is presently under investigation.

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