Phase Separation in the Two-Dimensional Bosonic Hubbard Model with Ring Exchange

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We show that soft-core bosons in two dimensions with a ring exchange term exhibit a tendency for phase separation. This observation suggests that the thermodynamic stability of normal Bose liquid phases driven by ring exchange should be carefully examined.

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Interest in ring exchange interactions in quantum many-body systems has a long history, both theoretically and experimentally [1]. Recently, the ring exchange interaction has been invoked in an effort to understand various aspects of high temperature superconductivity. While the Heisenberg model alone provides a rather accurate picture of magnetic excitations in the parent compounds of the cuprate superconductors [2], estimates of the magnitude of the ring exchange term are as high as one quarter of the exchange coupling [3–5] and it therefore has been of interest to understand how this term might modify magnetic properties [3,6–9]. Ring exchange interactions have also been suggested as a likely candidate to reconcile the properties of the underdoped pseudogap regime. The basic picture is that the ring exchange interaction can give rise to a new normal "Bose metal" phase at zero temperature in which there are no broken symmetries associated with superfluidity or charge density wave phases, and in which the compressibility is also finite [10].

With these motivations partly in mind, Sandvik *et al.* [11] studied the phase diagram of the two-dimensional spin-1/2 XY model with spin exchange interaction on a square lattice,

$$H = -J\sum_{\langle \mathbf{i}\mathbf{j}\rangle} B_{\mathbf{i}\mathbf{j}} - K\sum_{\langle \mathbf{i}\mathbf{j}\mathbf{k}\mathbf{l}\rangle} P_{\mathbf{i}\mathbf{j}\mathbf{k}\mathbf{l}}, \tag{1}$$

where

$$\begin{aligned} B_{\mathbf{i}\mathbf{j}} &= S_{\mathbf{i}}^{+}S_{\mathbf{j}}^{-} + S_{\mathbf{i}}^{-}S_{\mathbf{j}}^{+} = 2(S_{\mathbf{i}}^{x}S_{\mathbf{j}}^{x} + S_{\mathbf{i}}^{y}S_{\mathbf{j}}^{y}), \\ P_{\mathbf{i}\mathbf{j}\mathbf{k}\mathbf{l}} &= S_{\mathbf{i}}^{+}S_{\mathbf{j}}^{-}S_{\mathbf{k}}^{+}S_{l}^{-} + S_{\mathbf{i}}^{-}S_{\mathbf{j}}^{+}S_{\mathbf{k}}^{-}S_{l}^{+}, \end{aligned} \tag{2}$$

and $\langle \mathbf{i}\mathbf{j}\rangle$ denotes nearest neighbors and $\langle \mathbf{i}\mathbf{j}\mathbf{k}\mathbf{l}\rangle$ are sites at the corners of a plaquette. As is well known, for $K = 0$ this model is exactly equivalent to the hard-core bosonic Hubbard model, at half filling, with no interactions apart from the constraint on-site occupations, and it has only a superfluid phase. Sandvik *et al.* [11] studied the phase diagram as a function of K using the stochastic series expansion algorithm [12]. They found that as K increases, the superfluid density, ρ_s, decreases up to a critical value, K_c, where a phase transition takes place, and ρ_s goes to zero with long range order appearing in the momentum $(\pi, 0)$ and $(0, \pi)$ channels of the plaquette-plaquette correlation function. This indicates the existence of a phase in which plaquettes with large and small values of ring exchange alternate with a striped pattern across the lattice. This phase is also an incompressible insulator. For yet larger K, charge density wave order is established in which the site occupations are alternatingly large and small.

It is believed [10] that when the hard-core constraint is relaxed, the striped plaquette phase might evolve into a normal compressible conducting "Bose metal" in which none of the order parameters mentioned above is nonzero. This suggestion leads us to study here the phase diagram of the soft-core bosonic Hubbard model at half filling with ring exchange interaction,

$$\begin{aligned} H = &-t\sum_{\langle \mathbf{i},\mathbf{j}\rangle}(a_{\mathbf{i}}^{\dagger}a_{\mathbf{j}} + a_{\mathbf{j}}^{\dagger}a_{\mathbf{i}}) + U\sum_{\mathbf{i}} n_{\mathbf{i}}(n_{\mathbf{i}} - 1) \\ &- K\sum_{\langle \mathbf{i}\mathbf{j}\mathbf{k}\mathbf{l}\rangle}(a_{\mathbf{i}}^{\dagger}a_{\mathbf{j}}a_{\mathbf{k}}^{\dagger}a_{l} + a_{\mathbf{i}}a_{\mathbf{j}}^{\dagger}a_{\mathbf{k}}a_{l}^{\dagger}), \end{aligned} \tag{3}$$

where the destruction and creation operators satisfy $[a_{\mathbf{i}}, a_{\mathbf{j}}^{\dagger}] = \delta_{\mathbf{i}\mathbf{j}}$, $n_{\mathbf{i}} = a_{\mathbf{i}}^{\dagger}a_{\mathbf{i}}$ is the number operator at site $\mathbf{i}$ and U is the on-site interaction strength. We choose $t = 1$ to set the energy scale. For our quantum Monte Carlo (QMC) simulations we used the world line algorithm with four-site decoupling [13]. We verified our code for $K = 0$ by comparing with existing results for hard- and soft-core bosons with and without near and next near neighbor interactions. For the $K \neq 0$ case, we compared with the hard-core results of [11].

Before discussing results for the full many-body system, it is interesting to study the behavior of two bosons, since the formation of a bound state is closely related to the issue of phase separation. Ring exchange, like an attractive potential, favors proximity of two bosons, since the action of such a term is nonzero only when the two bosons live on the same plaquette. In Fig. 1 we show the average separation $\langle \Phi_0|r^2|\Phi_0\rangle$ of two bosons, normalized to the number of sites L^2 on an $L \times L$ lattice. As K

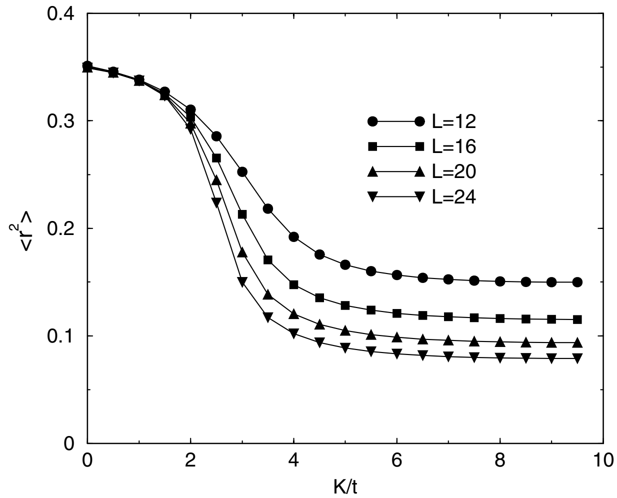

FIG. 1. Average ground state separation $\langle \Phi_0 | r^2 | \Phi_0 \rangle$ of two bosons, normalized to the system size, as a function of the magnitude of the ring exchange energy scale K. Here the hopping $t = 1$ and the soft-core repulsion $U = 12$. At small K, the normalized separation is independent of lattice size, indicating that the two bosons are spread independently throughout the lattice. At larger K the bosons prefer smaller separation to optimize the ring exchange energy, indicating the formation of a bound state.

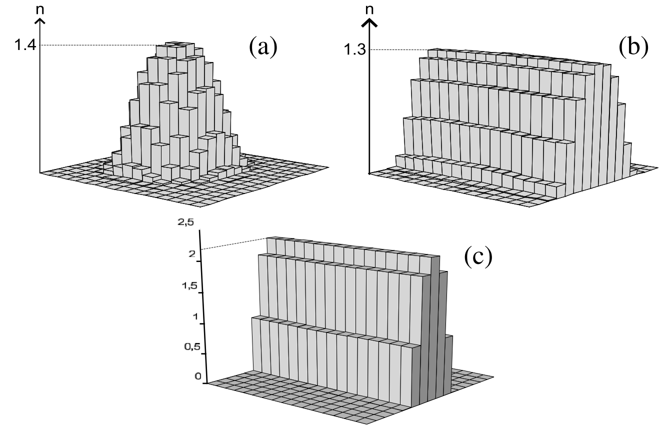

FIG. 2. Typical QMC results for the average density distribution in the phase separated region. Panel (a): $U = 4$, $K = 2.5$, and $\rho = 50/256$; panel (b): $U = 4$, $K = 2.5$, and $\rho = 128/256$ (half filling); panel (c): $U = 4$, $K = 10$, and $\rho = 128/256$ (half filling). At half filling, a stripe across our periodic boundary condition lattice maximizes the number of occupied plaquettes, and hence minimizes the ring exchange energy. As K increases, the stripe becomes narrower, reflecting the tendency of K to localize the bosons.

increases, there is a crossover at $K \approx 3$ from a regime where the boson separation grows linearly with system size, so that the normalized separation is size independent, to one in which the boson separation does not grow with system size.

While we do not show the associated data, plots of $\langle \Phi_0 | r^2 | \Phi_0 \rangle$ for different soft-core repulsion U reveal that the average separation is insensitive to the value of U. This will obviously be true in the unbound regime at small K, since the density is so dilute. It is less clear that this should be so in the bound regime at large K. However, as can be seen from the data in Fig. 1, the radius of the bound state is several lattice spacings ($\langle r^2 \rangle \propto 0.1L^2$ whence $r \propto 0.3L$), so here too the effect of U is expected to be relatively small.

Figure 1 suggests that there might be a tendency for ring exchange to cause the bosons to clump together, and, in an extreme scenario, to undergo phase separation. However, at densities higher than the dilute two boson case, this effect is opposed by the repulsion U. The focus of this Letter is to examine this competition and determine the phase diagram of the soft-core case as a function of both U and K at half filling.

The most straightforward indication of phase separation comes from a real space image of the boson density during the course of a simulation. Panels (a) and (b) of Fig. 2 show the average density distribution [14] for $L = 16$, $U = 4$, and$K = 2.5$. We see indications that the bosons undergo phase separation: At less than quarter filling [panel (a)] the bosons clump together into a compact region of high density. At densities above quarter filling, on a lattice with periodic boundary conditions, the number of occupied plaquettes is largest (and hence the ring exchange energy is most negative) for a configuration where the bosons stretch out in stripe across the lattice [panel (b)]. Panel (c) shows the density distribution at large K, beyond the quantum critical point identified in Ref. [11].

We will now demonstrate that phase separation is characteristic of a large portion of the K-U phase diagram by examining the density-density correlation function and its associated structure factor, fixing U (or K) and scanning K (or U). As Fig. 2 illustrates, if the bosons phase separate, they may form a structure in which a set of contiguous sites of about half the system size will have appreciable boson occupation. The other half of the lattice is essentially empty. Therefore, if one examines the structure factor of the density-density correlation function,

$$S(k_x, k_y) \equiv \frac{1}{L^2} \sum_{\mathbf{r}} C(\mathbf{r}) \mathrm{e}^{-i\mathbf{r}.\mathbf{k}} \tag{4}$$

with

$$C(\mathbf{r}) = \frac{1}{L^2} \sum_{\mathbf{r}'} \langle n(\mathbf{r}') n(\mathbf{r} + \mathbf{r}') \rangle, \tag{5}$$

one should observe a peak in S at small momentum, e.g., $(2\pi/L, 0)$, $(0, 2\pi/L)$, or $(2\pi/L, 2\pi/L)$ depending on the precise orientation of the clump [15]. By looking at the sum of the density structure factor at these three smallest momentum values we are sensitive to phase separation regardless of whether it occurs in a puddle of roughly circular shape [Fig. 2(a)] or in some more elongated pattern [Fig. 2(b)]. Figure 3 clearly shows this behavior:

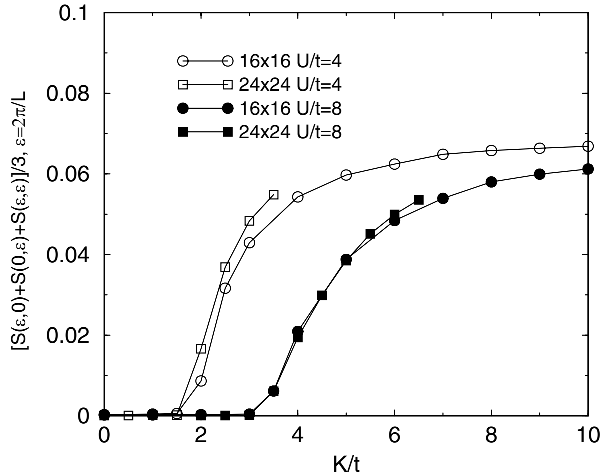

FIG. 3. The average structure factor, $[S(2\pi/L, 0) + S(0, 2\pi/L) + S(2\pi/L, 2\pi/L)]/3$ versus K. We see a sharp increase in S at $K = 2$ for $U = 4$ and $K \approx 4$ for $U = 8$. The simulations were done for $\beta = 8$ and $\rho = 0.5$.

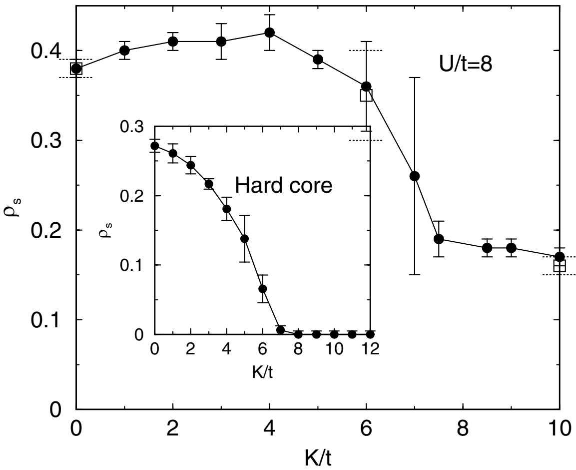

FIG. 4. The superfluid density as a function of K for $U = 8$. ρ_s remains finite in the phase separated region, indicating that the bosons are delocalized across the clump of occupied sites. Solid circles are 16×16 lattices and open squares are 24×24 lattices. Inset: The hard-core limit for which, instead, $\rho_s \to 0$ at large K.

For $K < 2$ at $U = 4$, S is very small at the relevant momenta. For $K > 2$, phase separation sets in. Data for 16×16 and 24×24 lattices are shown and their agreement indicates that this phase separation is not a finite lattice effect. The critical value of K grows roughly linearly with U.

It is also interesting to understand the behavior of the superfluid density ρ_s. One does not necessarily expect ρ_s to vanish when phase separation occurs. In fact, as is well known, $\rho_s \neq 0$ for the soft-core boson Hubbard model at all fillings, including commensurate density, if U is sufficiently small. Similarly, here it is possible that ρ_s can survive in the dense region of the phase separated lattice [16,17]. Our simulations show that when phase separation first occurs, the populated region forms a band that spans the whole system. The bosons may then delocalize along that band, maintaining an (anisotropic) superfluid density. We have found that when the bosons form such a band, the plaquette-plaquette structure factor is also anisotropic and has long range correlations along the direction of the band. As K is increased further, the populated region of the lattice takes the form of an island. In such a case, the system may not be considered a superfluid in that one cannot establish superflow across the system. However, the bosons may still be delocalized over the extent of the island [18]. Figure 4 shows ρ_s versus K [19].

By making scans like those of Fig. 3 at several values of U, we construct the phase diagram which we show in Fig. 5. Above the solid line, the system undergoes phase separation.

How do these results connect with the previous studies in the hard-core limit? There we know that a phase transition occurs at $K_c/J \approx 7.9$ from a superfluid to a striped plaquette phase [11]. We show in Fig. 6 the behavior of the boson density ρ as a function of chemical potential μ in this hard-core limit. The jump in μ across half filling $\rho = 0.5$ shows that the plaquette ordered phase has a gap to the addition of bosons (“charge excitations”), a result which is in agreement with Sandvik *et al.* [11]. Melko *et al.* report jumps in the magnetization and superfluid density at large K which similarly indicate a first order transition [20]. In Fig. 6, the slope of the ρ versus μ is the compressibility κ. Consequently, if this curve “bends backwards,” the system is thermodynamically unstable and undergoes phase separation [21,22]. While the data are not conclusive, we do see hints of an instability. For $|\rho - 0.5| > 0.008$, the slope is finite and corresponds to a normal superfluid. For $|\rho - 0.5| < 0.008$, the slope is either very large, or perhaps negative, indicative of phase separation.

So far we have addressed mostly the half-filled case, and lower densities. It is of course of interest to examine

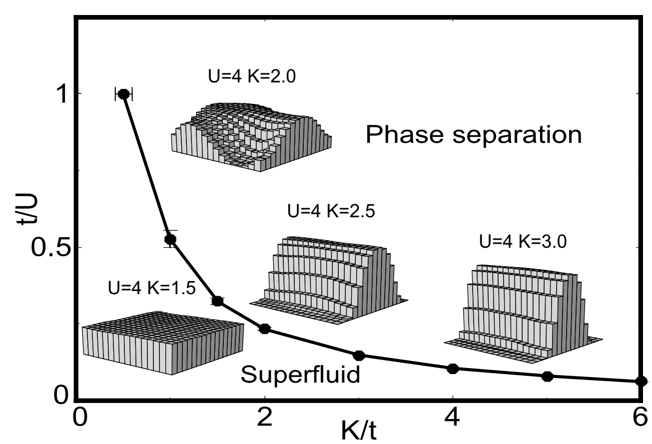

FIG. 5. The phase diagram of the half-filled Bose-Hubbard model in the U-K plane. Below the solid line, the system is superfluid while above the line it phase separates; see text.

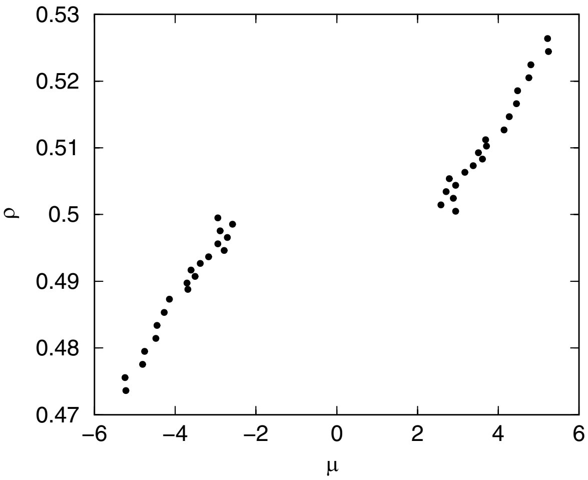

FIG. 6. The boson density, ρ, versus the chemical potential, μ, for hard-core bosons at $K = 10$. The jump in μ at $\rho = \frac{1}{2}$ is associated with the nonzero gap in the phase with long range striped plaquette order. There are indications of a region of negative compressibility $\kappa = \partial\rho/\partial\mu$ immediately adjacent to the gapped phase.

higher fillings where the effect of U will be expected to discourage phase separation. We have done simulations for $\rho = 1$ and $\rho = 1.5$ and found in both cases that despite the higher densities, phase separation still sets in above a critical value $K > K_c$ of the ring exchange energy scale. This result is not so surprising since the on-site repulsion U and the ring exchange term both scale with density as ρ^2. (Each of the four ring exchange creation/destruction operators picks up a factor of $\approx \sqrt{n}$ when acting on a site with occupation n.)

In conclusion, we have shown that a sufficiently large ring exchange energy can lead to a thermodynamic instability and phase separation. We determined the critical K as a function of the soft-core repulsion U for a half-filled lattice and found roughly $K_c \approx U/2$. We conclude that the soft-core boson Hubbard model does not exhibit a normal Bose metal phase, but instead undergoes phase separation. This phase separation also takes place when the hopping parameter vanishes, $t = 0$, a limit examined for the quantum phase model in Ref. [10] but which did not find phase separation.

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[1] D. J. Thouless, Proc. Phys. Soc. London **86**, 893 (1965).
[2] E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).
[3] R. Coldea *et al.*, Phys. Rev. Lett. **86**, 5377 (2001).
[4] A. A. Katanin and A. P. Kampf, Phys. Rev. B **66**, 100403(R) (2002).
[5] E. Muller-Hartmann and A. Reischl, Eur. Phys. J. B **28**, 173 (2002).
[6] M. Roger and J. M. Delrieu, Phys. Rev. B **39**, 2299 (1989).
[7] Y. Honda *et al.*, Phys. Rev. B **47**, 11 329 (1993).
[8] J. Lorenzana *et al.*, Phys. Rev. Lett. **83**, 5122 (1999).
[9] M. Matsuda *et al.*, Phys. Rev. B **62**, 8903 (2000).
[10] A. Paramekanti *et al.*, Phys. Rev. B **66**, 054526 (2002).
[11] A.W. Sandvik *et al.*, Phys. Rev. Lett. **89**, 247201 (2002).
[12] A.W. Sandvik and J. Kurkijarvi, Phys. Rev. B **43**, 5950 (1991).
[13] E. Loh *et al.*, Phys. Rev. B **31**, 4712 (1985).
[14] The data shown in Fig. 2 are not a snapshot of a single configuration, but an average over a whole simulation run. In principle for extremely long simulations, the puddle of occupied sites will gradually move around the whole lattice and give a uniform density, $\rho_{\mathbf{i}} = \frac{1}{2}$ on every site $\mathbf{i}$. In practice, such motion of the large collection of bosons is extremely slow.
[15] When U is reasonably large and all sites are either empty or singly occupied, the structure factor at $S[k = (0, 0)]$ is just the density.
[16] G. G. Batrouni *et al.*, Phys. Rev. Lett. **65**, 1765 (1990).
[17] M. P. A. Fisher *et al.*, Phys. Rev. B **40**, 546 (1989).
[18] There are interesting analogies between this sort of "local" superfluidity and the behavior of bosons in a confining potential. See, e.g., M. Greiner *et al.*, Nature (London) **415**, 39 (2002).
[19] We measure ρ_s through an extrapolation of the imaginary time dependence of an appropriate current-current correlation function. For details, see Ref. [16].
[20] R. G. Melko, A.W. Sandvik, and D. J. Scalapino, Phys. Rev. B **69**, 100408 (2004).
[21] G. G. Batrouni and R. T. Scalettar, Phys. Rev. Lett. **84**, 1599 (2000).
[22] F. Hébert *et al.*, Phys. Rev. B **65**, 014513 (2002).