

Dimer State of Spin-1 Bosons in an Optical Lattice

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We consider spin-1 bosons, such as ^{23}Na , trapped in an optical lattice, in the regime of one particle per site. We argue that the ground state is expected to be the dimer phase in one, two, or three dimensions, thus realizing a state that has so far been studied only theoretically.

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Tremendous progress has been made recently on trapping and cooling atoms. Greiner *et al.* [1] have succeeded in observing the bosonic Mott-Hubbard transition. In the experiment, the bosons involved are ^{87}Rb atoms and the lattice potential is provided by the standing waves of three orthogonal laser beams. For weak lattice potentials, the system remains a superfluid (Bose-Einstein condensate). When the lattice potential is increased beyond a critical strength, the tunneling rate for atoms between the different wells becomes weak compared with the repulsion for two atoms residing in the same well; the system enters into the Mott regime. In this case the number of atoms in each well is essentially fixed at an integer value (here, 1). Phase coherence and hence superfluidity is lost [2,3].

The ^{87}Rb atoms in the experiment of Greiner *et al.* [1] were hyperfine spin-polarized. In this Letter, we discuss the interesting physics that can be realized if ^{23}Na atoms are employed instead of ^{87}Rb and if the atoms are not polarized. Bose-Einstein condensation of unpolarized ^{23}Na has already been achieved by the MIT group [4]. The ^{23}Na atoms have hyperfine spin (hereafter simply “spin”) 1 in their lower energy manifold, and the interaction among them is antiferromagnetic [4,5] (in contrast to ^{87}Rb , where it is ferromagnetic). We confine ourselves to the Mott regime where there is essentially one atom per cell and discuss the arrangement of the hyperfine spin states (or, more precisely, the projections) for the atoms in this lattice. We argue that the ground state is expected to be the dimer phase in one, two, or three dimensions. This is in contrast with spin-1 electronic systems where the effective Hamiltonian is the antiferromagnetic Heisenberg model. There in one dimension, the system would be in the Haldane phase, whereas long range Néel order is expected to be present in two and three dimensions [6].

Consider then a (cubic) lattice formed by three orthogonal laser beams, with effective potential for the ^{23}Na atoms of the form $V(x, y, z) = V_x \sin^2(kx) + V_y \sin^2(ky) + V_z \sin^2(kz)$, where $k \equiv \frac{2\pi}{\lambda}$ is the wave vector of the lasers. The strength $V_{x,y,z}$ of the sinusoidal potentials is proportional to the intensities of the laser beams and can be adjusted separately. As in Ref. [3] we assume that only one orbital state is involved for each well. The effective Hamiltonian for our system can be written

in a Bose-Hubbard form [3] (generalized to bosons with spin). The hopping matrix elements $t_{x,y,z}$ are dependent on the directions of hopping. The order of magnitude of t_x is given by $|t_x| \sim (E_R V_x)^{1/2} e^{-2(V_x/E_R)^{1/2}}$, where $E_R \equiv \hbar^2 k^2 / 2m$ and m is the mass of the atoms. Though we are considering one particle per well, the system can exist in excited states where the wells are multiply occupied. We confine ourselves to the regime where these energies are large compared with the hopping energies $|t_{x,y,z}|$. Thus we need consider only those excited states with at most two particles per well. The extra energy is described by the Hubbard repulsion U_S which depends on the total spin S of the two particles involved. $U_S \sim (E_R V_x V_y V_z)^{1/4} k a_S$, where a_S is the scattering length in total spin $S = 0, 2$ channel. Excited states with $S = 1$ are not allowed due to the identity of the bosons with one orbital state per well.

We next construct, in the standard manner, the effective Hamiltonian for the spins in the subspace of exactly one particle per well, assuming $|t| \ll U_{0,2}$ (\ll the excited energies $\sim (E_R V_{x,y,z})^{1/2}$ of higher orbital states in the wells: this latter inequality is typically satisfied since $a_S \ll \lambda$ [3]). Let us first consider two sites, labeled by 1 and 2. For $t \rightarrow 0$ the energy is independent of the spin configurations. For finite but small t , we can perform perturbation in t . It is convenient to classify the states according to the total spin S_{tot} of the two sites. To second order in t , the energy is lowered due to hopping by the amount $-4t^2/U_{0,2}$ for total spin $S_{\text{tot}} = 0, 2$ [7] but zero for total spin 1. With $\mathbf{S}_{1,2}$ the spin-1 operators for sites 1, 2 and using $\mathbf{S}_1 \cdot \mathbf{S}_2 = -2, -1, 1$ for $S_{\text{tot}} = 0, 1, 2$, respectively, we find that the effective Hamiltonian can be written as

$$\hat{H}_{12} = \epsilon + J(\mathbf{S}_1 \cdot \mathbf{S}_2) + K(\mathbf{S}_1 \cdot \mathbf{S}_2)^2, \quad (1)$$

where $J = -\frac{2t^2}{U_2}$, $K = -\frac{2}{3}\frac{t^2}{U_2} - \frac{4}{3}\frac{t^2}{U_0}$, and $\epsilon = J - K$. For ^{23}Na atoms, $U_2 > U_0 > 0$ [4,5,8]; hence $K < J < 0$. As a matter of fact, since $a_2 \approx 52a_B$ and $a_0 \approx 46a_B$ (here $a_B = \text{Bohr radius}$), $(U_2 - U_0)/U_0 = (a_2 - a_0)/a_0 \ll 1$. Hence $|J| \sim |K|$ and $|J - K| \ll |K|$ or $|J|$. (For ^{87}Rb , $U_0 > U_2 > 0$; in that case, $J < K < 0$.)

Thus the Hamiltonian (1) differs significantly from the Heisenberg one [9] familiar in ionic crystals. There, although the total spin of an ion can be larger than $1/2$

and hence $(\mathbf{S}_1 \cdot \mathbf{S}_2)$ and $(\mathbf{S}_1 \cdot \mathbf{S}_2)^2$ are independent operators, the Hamiltonian can, to a good approximation (ignoring spin-orbit interactions, etc.), be written simply as $\hat{H}_{Hei} = J(\mathbf{S}_1 \cdot \mathbf{S}_2)$. For perturbation to second order in the hopping of electrons between the two ions, there cannot be terms in $\mathbf{S}_1 \cdot \mathbf{S}_2$ with powers higher than 1 in the effective Hamiltonian, since two hoppings of electrons with spin 1/2 can change only the z component of the spin of an ion by at most 1, whereas, e.g., $(\mathbf{S}_1 \cdot \mathbf{S}_2)^2$ consists of terms which can change that component by ± 2 . Higher order perturbation in hopping can give rise to terms higher powers in $(\mathbf{S}_1 \cdot \mathbf{S}_2)$ but only with much smaller coefficients when hopping is small compared with the Hubbard repulsion.

Another peculiar fact about Hamiltonian (1) is also apparent. If the spins were classical vectors, $K < J < 0$ would require that the two spin vectors be *parallel* in the lowest energy state. However, since $U_2 > U_0 > 0$, $E_{S=0} < E_{S=2}$ and hence the spins on the two sites actually prefer to be *antiferromatically* correlated. The strong quantum mechanical nature of the spins will be of significance below.

Let us now begin with the case where $V_x \ll V_{y,z}$, so that one can ignore couplings along y and z directions. We thus have a collection of one-dimensional spin-1 chains. Ignoring the ϵ term in Eq. (1) not of relevance below, our effective Hamiltonian is thus (for one chain)

$$\begin{aligned} \hat{H} &= \sum_l [J(\mathbf{S}_l \cdot \mathbf{S}_{l+1}) + K(\mathbf{S}_l \cdot \mathbf{S}_{l+1})^2] \quad (2) \\ &\equiv \sqrt{J^2 + K^2} \sum_l [\cos\gamma(\mathbf{S}_l \cdot \mathbf{S}_{l+1}) + \sin\gamma(\mathbf{S}_l \cdot \mathbf{S}_{l+1})^2], \quad (3) \end{aligned}$$

where the sum is over the site labels l and the second relation defines γ . For $K < J < 0$ $-\frac{3\pi}{4} < \gamma < -\frac{\pi}{2}$. If $|U_2 - U_0| \ll U_0 \leq U_2$ as for ^{23}Na , $\gamma \rightarrow -\frac{3\pi}{4}$, whereas if $U_2 \gg U_0 > 0$, $\gamma \rightarrow -\frac{\pi}{2}$. (For $J < K < 0$ as in ^{87}Rb , $-\pi < \gamma < -\frac{3\pi}{4}$.)

A Hamiltonian of the form in (3) has been studied before, with most efforts in the region where the ground state is expected to be in the Haldane phase ($-\frac{\pi}{4} < \gamma < \frac{\pi}{4}$) [10]. There have been many fewer studies in the range of γ of relevance here. Though it is agreed that for γ near $-\frac{\pi}{2}$ the ground state should be in a dimer state [11], results for other γ 's have been somewhat controversial, especially near $-\frac{3\pi}{4}$. In particular, Chubukov [12] suggested that there is a critical γ_c ($-\frac{3\pi}{4} < \gamma_c < -\frac{\pi}{2}$) such that the dimer state is unstable for $\gamma < \gamma_c$, where instead the nematic phase should exist. A later study [13] claims otherwise. Recently Demler and Zhou [14] considered possible ground states for spin-1 bosons in optical lattice as in the present Letter but did not include the dimer state in their discussion.

We first readdress this issue by variational ansatz. We denote the three possible spin projection states at a given

site by $|+\rangle$, $|0\rangle$, and $|-\rangle$ and write the wave functions by specifying these states at each site. The nematic phase is given by

$$\Psi_{\text{nem}} = |\dots 0000 \dots\rangle. \quad (4)$$

This state is in direct analogy with the corresponding ‘‘polar’’ phase [5] in the bulk. This state has $\langle \mathbf{S}_l \rangle = 0$ and $\langle S_{l,x}^2 \rangle = \langle S_{l,y}^2 \rangle \neq \langle S_{l,z}^2 \rangle$ [15]. For the dimer state, the usually employed ansatz is [10,12]

$$\Psi_{\text{dimer}} = \dots \Psi_{12} \Psi_{34} \dots, \quad (5)$$

where

$$\Psi_{12} = \frac{1}{\sqrt{3}}(|+-\rangle + |-+\rangle - |00\rangle)_{12} \quad (6)$$

is a singlet ($S_{\text{tot}} = 0$) pair formed by sites 1 and 2 (the subscripts label the sites). This state is shown schematically in Fig. 1(a). [There is another state degenerate with (5) with all pairs shifted by one lattice site.] In the *non-interacting* spin-wave approximation, the $S = 2$ modes become unstable at $\gamma_c = -\pi + \tan^{-1} \frac{9}{5} \approx -0.66\pi$. Chubukov [12] then concludes that for $-\frac{3\pi}{4} < \gamma < \gamma_c < -\frac{\pi}{2}$ (with γ_c possibly renormalized), the dimer state is unstable and further speculates that the correct ground state should be the nematic state.

Here we revisit this question of stability by studying an improved variational ansatz. We write again Eq. (5) but with the pair wave function given by

$$\Psi_{12} = \frac{1}{\sqrt{2 + |\zeta|^2}}(|+-\rangle + |-+\rangle - \zeta|00\rangle)_{12}, \quad (7)$$

etc., with ζ our variational parameter. If $\zeta = 1$, our ansatz reduces to that in Eq. (6). Notice that if $|\zeta| \rightarrow \infty$, then $\Psi_{\text{dimer}} \rightarrow \Psi_{\text{nem}}$. It is simple to evaluate the expectation values of \hat{H} . We find, with $\zeta = |\zeta|e^{i\phi}$,

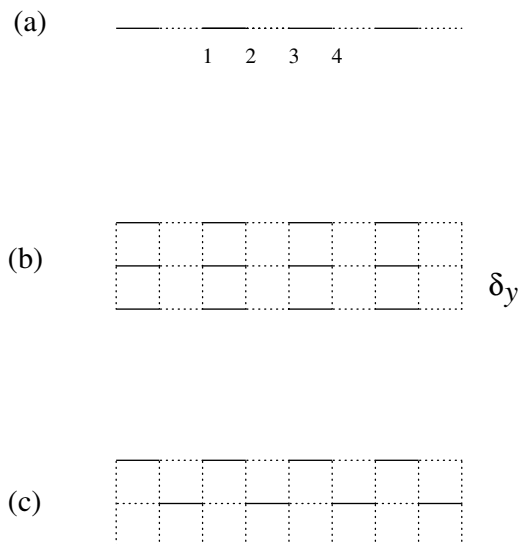


FIG. 1. Dimers. The thick lines represent the pairs such as Eq. (7).

$$\langle \hat{H}_{12} \rangle = \frac{2}{2 + |\zeta|^2} \{ -(1 + 2|\zeta| \cos \phi) \cos \gamma + (3 + 2|\zeta| \cos \phi + |\zeta|^2) \sin \gamma \} \quad (8)$$

for the bond (the part of \hat{H}) between sites 1 and 2, and

$$\langle \hat{H}_{23} \rangle = \frac{2}{[2 + |\zeta|^2]^2} \{ (3 + 2|\zeta|^2 + |\zeta|^4) \sin \gamma \} \quad (9)$$

for the bond between sites 2 and 3. The energy E per site is thus given by

$$E_{\text{dimer}} = \frac{1}{2} [\langle H_{12} \rangle + \langle H_{23} \rangle]. \quad (10)$$

The ϕ dependent part of E arises only from $\langle \hat{H}_{12} \rangle$ and is proportional to $(\sin \gamma - \cos \gamma) \cos \phi$ with a positive coefficient. For our region of γ , $(\sin \gamma - \cos \gamma) < 0$; hence the energy is minimized at $\phi = 0$, i.e., ζ real and positive. Henceforth we put $\phi = 0$ and restrict $0 \leq \zeta \leq \infty$.

By expanding Eq. (10) near $\zeta = 1$, we find

$$E_{\text{dimer}} = (-\cos \gamma + \frac{8}{3} \sin \gamma) + \frac{1}{3} (\cos \gamma - \frac{5}{9} \sin \gamma) \times (\zeta - 1)^2 + \dots \quad (11)$$

Thus $\zeta = 1$ is a relative energy minimum provided $\gamma > \gamma_c \equiv -\pi + \tan^{-1} \frac{9}{5} \approx -0.66\pi$ defined above. This result is in accordance with the discussions following Eq. (6). However, we here do not interpret this as an instability of the dimer state, but rather that a better or more generalized ansatz is required as is done here. E_{dimer} as a function of ζ is plotted in Fig. 2(a). For $\gamma < \gamma_c$ we find that the minimum energies occur at $1 < \zeta < \infty$. Thus though the pair wave function differs from the singlet state, the system is still dimerized. (It can be seen easily that $\langle H_{12} \rangle \neq \langle H_{23} \rangle$ provided $\zeta \neq \infty$.) ζ increases without limit when γ decreases towards $-\frac{3\pi}{4}$. To examine the stability of the dimer state versus the nematic phase, we expand Eq. (10) as a function of $\eta = \zeta^{-1}$ near $\zeta = \infty$ ($\eta = 0$). We find

$$E_{\text{dimer}} = 2 \sin \gamma + 2(\sin \gamma - \cos \gamma) \eta + \dots, \quad (12)$$

where the ellipsis means terms of order η^2 or higher. Note that the energy of the nematic phase is given by putting $\eta = 0$; i.e., $E_{\text{nem}} = 2 \sin \gamma$. Thus the nematic phase is never stable for our γ region of interest, since $(\sin \gamma - \cos \gamma) < 0$. The behavior of E_{dimer} as a function of ζ for large ζ is plotted in Fig. 2(b).

Since $(\sin \gamma - \cos \gamma)$ changes sign at $\gamma = -\frac{3\pi}{4}$, the nematic phase (4) is more stable than the dimer state (7) for $\gamma < -\frac{3\pi}{4}$. However, for $-2\pi < \gamma < -\frac{3\pi}{4}$ the ferromagnetic state

$$\Psi_{\text{ferro}} = |\dots + + \dots\rangle \quad (13)$$

[or any other state obtained by applying the lowering operator $\mathbf{S}_{\text{tot},-}$ arbitrary (limited only by twice the number of sites) number of times], with energy per site $E_{\text{ferro}} = \cos \gamma + \sin \gamma$, becomes more stable than the nematic

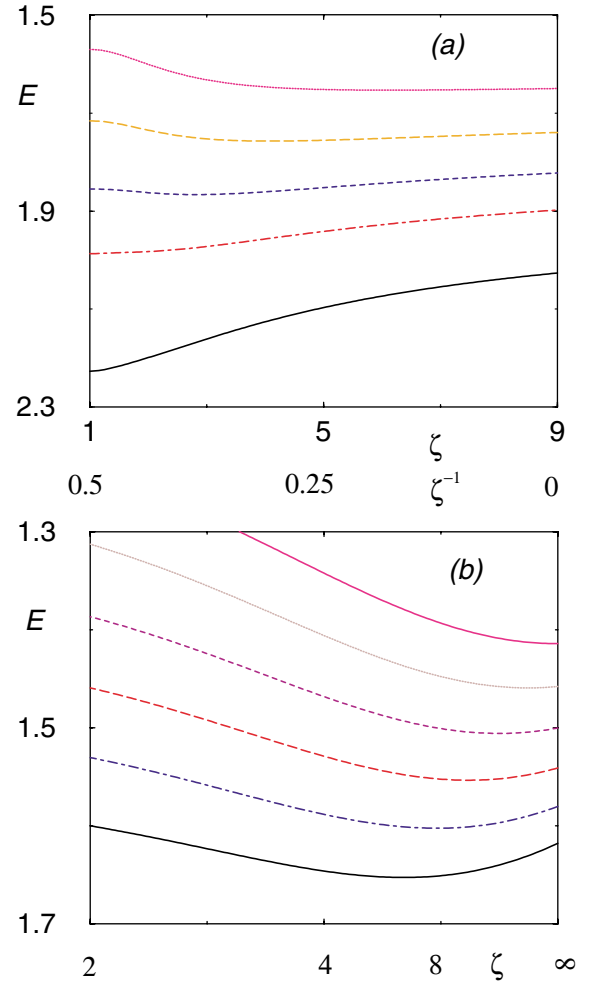


FIG. 2 (color online). The energy E_{dimer} per site for the ansatz wave function (5) and (7) as a function of the parameter ζ . The lines are for, from bottom to top and in units of π , (a) $\gamma = -0.6, -0.64, -0.66, -0.68, \text{ and } -0.7$; (b) $\gamma = -0.7, -0.71, -0.72, -0.73, -0.74, \text{ and } -0.75$.

phase. Thus the nematic phase has no regime of stability for the Hamiltonian in Eq. (3).

The ansatz wave function (7) does not have definite total spin S_{tot} unless $\zeta = 1$. Moreover, the wave function should be improved by including spin-wave fluctuations. However, we expect that the projection onto definite S_{tot} [16] and correcting the states with correlations between pairs will not change qualitatively the picture given above.

Consider now finite V_y but still $V_z \rightarrow \infty$. We thus now have a collection of two-dimensional spin-1 lattices. In general the tunneling matrix elements along the x and y directions, hence the strength of the effective spin interactions, can be unequal. The effective spin Hamiltonian for our lattice is of the same form as Eq. (2), except that we need two labels (l_x, l_y) for each lattice point, and the interactions can be between nearest neighbor pairs along both the x and y directions, with interaction constants

$J_{x,y}, K_{x,y}$. With $t_y = \delta_y^{1/2} t_x$, we have $J_y = \delta_y J_x$ and $K_y = \delta_y K_x$. Without loss in generality we can take $0 < \delta_y < 1$. Since the excitations of the dimer phase are gapped [10,12], we expect that the dimer phase is stable at least for small δ_y . It is straightforward to generalize our ansatz (5) and (7) above to the two-dimensional case, with schematic wave function as shown in Fig. 1(b). For each site, there is one “strong” bond along the x direction with energy as in Eq. (8), one “weak” bond also along x with energy as in Eq. (9), and two weak bonds along the y direction with total energy given by $2\delta_y$ times Eq. (9). From this total energy, we can see that our discussions for one dimension is basically unaffected. In particular, since Eq. (9) does not contribute any term linear in $\eta = \zeta^{-1}$, the discussions below Eq. (12) is qualitatively unchanged. Within our ansatz, the nematic phase is still always unstable towards the dimer phase for any $-\frac{3\pi}{4} < \gamma < -\frac{\pi}{2}$ and $0 \leq \delta_y < 1$.

Under the above variational ansatz, the states are degenerate with respect to the spatial arrangements of the pairs. In particular, the ansatz wave functions corresponding to Figs. 1(b) and 1(c), with the pairs forming a rectangular and a triangular lattice, respectively, are completely degenerate in energy. However, this degeneracy will be lifted once spin-wave fluctuations are taken into account. For $\zeta = 1$ the spin-wave spectrum can be found as in Ref. [12]. There are three $S = 1$ modes and five $S = 2$ modes, with dispersions $\omega_1(\vec{q}) = X_1[1 - \frac{2Y_1\nu(\vec{q})}{X_1}]^{1/2}$ and $\omega_2(\vec{q}) = X_2[1 + \frac{2Y_2\nu(\vec{q})}{X_2}]^{1/2}$, where $X_1 \equiv \cos\gamma - 3\sin\gamma$, $X_2 \equiv 3(\cos\gamma - \sin\gamma)$, $Y_1 \equiv \frac{2}{3}(2\cos\gamma - \sin\gamma)$ and $Y_2 \equiv \frac{2}{3}\sin\gamma$. (X_1, X_2 are positive in our region of γ .) Denoting the distance between sites by $a \equiv \pi/k$, for the rectangular lattice, $\nu^{\text{rect}}(\vec{q}) = \cos(2q_x a) + 2\delta_y \cos(q_y a)$, and for the triangular lattice, $\nu^{\text{tri}}(\vec{q}) = \cos(\vec{q} \cdot \vec{a}) + \delta_y \cos[\vec{q} \cdot (\vec{a} - \vec{b})] + \delta_y \cos(\vec{q} \cdot \vec{b})$. Here $\vec{a} \equiv (2a)\hat{x}$ and $\vec{b} \equiv a(\hat{x} + \hat{y})$ are the lattice vectors for our triangular lattice. The correction to the energy from the spin waves is given by $\Delta E = \sum_{\vec{q}} \frac{3}{2} [\omega_1(\vec{q}) - (X_1 - Y_1\nu(\vec{q}))] + \frac{5}{2} [\omega_2(\vec{q}) - (X_2 + Y_2\nu(\vec{q}))]$. Evaluating this energy for small δ_y , we find that the rectangular lattice has lower energy. Assuming that the lattice type applies for the entire region of γ of interest here, we conclude that the ground state for two dimensions should be as shown in Fig. 1(b). Similar considerations suggest that, for three dimensions, the lattice for ground state should be tetragonal.

The distinguishing property of the dimer state is the doubling of the unit cell, while the spins do not have

long range order. Thus the periodicity of the excitations with period $\Delta q = \pi/a = k = 2\pi/\lambda$ for a certain direction (x above) would be the signature of the dimer state. These excitations can, in principle, be detected by scattering.

In conclusion, we have pointed out that the trapped bosonic atoms provide the opportunity to realize a quantum mechanical state predicted in theory of quantum magnetism so far not tested experimentally. Obtaining this state in the laboratory would further widen the playground for quantum many-body systems.

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