Controlling two-species Mott-insulator phases in an optical lattice to form an array of dipolar molecules

M. G. Moore† and H. Sadeghpour†
ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138

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We consider the transfer of a two-species Bose-Einstein condensate into an optical lattice with a density such that that a Mott-insulator state with one atom per species per lattice site is obtained in the deep lattice regime. Depending on collision parameters the result could be either a ‘mixed’ or a ‘separated’ Mott-insulator phase. Such a ‘mixed’ two-species insulator could then be photo-associated into an array of dipolar molecules suitable for quantum computation or the formation of a dipolar molecular condensate. For the case of a $^{87}\text{Rb}-^{41}\text{K}$ two-species BEC, however, the large inter-species scattering length makes obtaining the desired ‘mixed’ Mott insulator phase difficult. To overcome this difficulty we investigate the effect of varying the lattice frequency on the mean-field interaction and find a favorable parameter regime under which a lattice of dipolar molecules could be generated.

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In a recent experiment [1] a quantum phase transition from a superfluid (SF) to a Mott insulator (MI) state was observed by varying the depth of a three-dimensional optical lattice [2] superimposed onto a trapped Bose-Einstein condensate (BEC) of $^{87}\text{Rb}$ atoms. This experiment highlights a growing trend in ultracold atomic physics whereby condensates are no longer the direct object of study, but serve mainly as well-controlled initial state for the preparation of more exotic highly-correlated many-body states. In addition to providing new insight into fundamental phenomena of condensed-matter physics, MI states are expected to have important applications in Heisenberg-limited atom-interferometry [3] and quantum computing [4,5].

Another interesting application of a Mott insulator was recently proposed by Jaksch and coworkers: employing a Mott insulator state as an intermediate stage in the generation of a BEC of molecules [6]. Molecules would be formed via stimulated Raman photoassociation of a Mott insulator with two atoms per lattice site, resulting in a molecular MI with unit filling factor which could then be ‘melted’ to form a molecular BEC. The primary advantage of this approach is that during photoassociation the resulting molecules would be completely isolated from collisions with other atoms and/or molecules. This is a distinct advantage over photoassociation in the superfluid phase [7,8], as photoassociation strongly favors the creation of vibrationally excited molecules due to favorable Franck-Condon transition from the continuum-which tend to undergo inelastic collisions in the presence of other atoms and/or molecules. The resulting release of vibrational quanta leads to loss of both particles from the trap. By isolating each vibrationally excited molecule in a lattice site, it should be possible to stimulate the molecules into the ground rotational-vibrational state before ‘melting’ the Mott insulator, thus eliminating this collisional loss mechanism.

In this Letter, we extend this idea to the formation of a lattice of two-species Mott insulator phase with precisely one atom per species per lattice site. This scheme would require a two-species Bose-Einstein condensate as a starting point, as has been recently demonstrated with $^{87}\text{Rb}$ and $^{41}\text{K}$ [9,10]. The two-species condensate would then be adiabatically loaded into a three-dimensional optical lattice, resulting in the formation of either a ‘mixed’ or ‘separated’ MI state, corresponding to each lattice site containing different or identical species, respectively, see Fig. 1. The formation of a mixed phase turns out to be difficult for the case of a $^{87}\text{Rb}-^{41}\text{K}$ system, due to the large positive inter-species scattering length which favors the separated phase. We therefore concentrate on overcoming this difficulty by varying the lattice frequency, which exploits the difference in atomic resonance frequencies to control the relative densities of the two species.

Preparing a Mott insulator state of heteronuclear molecules by this approach would be an elegant method to obtain a lattice of dipolar molecules for use as a quantum computer [11]. In addition a rich variety of quantum phases

FIG. 1. Separated- (left) and mixed- (right) two-species Mott insulator phases in an optical lattice. The separated-phase MI is randomly distributed.
are predicted for the ground state of a lattice of dipolar bosons, including a supersolid phase \cite{12}. The lattice of dipolar molecules could then be ‘melted’ by adiabatically lowering the lattice potential, resulting in a condensate of dipolar bosons. Here the long-range dipole-dipole interactions should introduce interesting correlations effects in both the ground state and collective excitations of the BEC \cite{13-17}.

Others have considered the related problem of superfluid-insulator transitions for spinor (multicomponent) BECs \cite{18} in which the added freedom of superpositions of hyperfine states is included. One crucial difference between a two-species system and a spinor system is the fact that each atomic species will see a different optical potential, which is then the bosonic annihilation operator for species \( i \), and \( g_{ij} \) is the collision rate for collisions between species \( i \) and \( j \).

In order to compute the various interaction and tunneling rates we must first consider the dependence of the optical lattice potential on the intrinsic properties of each atomic species. The optical potential seen by species \( i \) can be expressed as

\[
V_i(r) = \frac{d_i^2}{(\omega_i - \omega_L)} U(r),
\]

where \( d_i \) and \( \omega_i \) are the dipole moment and resonance frequency of species \( i \), respectively, \( \omega_L \) is the laser frequency of the optical field, and \( U(r) = -|E(r)|^2 / \hbar \) is the species-independent intensity of the optical field. Assuming that the sign of detuning is the same for both species, in which case the potential minima will coincide, we can expand around the potential minima according to \( U(r) = u_0 + \frac{1}{3} \sum_\mu u_\mu r_\mu^2 \), giving

\[
V_i(r) = \frac{1}{2} \frac{d_i^2}{(\omega_i - \omega_L)} \sum_\mu u_\mu r_\mu^2,
\]

where we have dropped the constant term consistent with a transformation to a rotating frame which leaves the Hamiltonian \( 1 \) unchanged.

Following the expansion in \( 3 \), we make a Gaussian approximation for the lowest energy Wannier modes of a lattice site for species \( i \) as

\[
\phi_i(r) = \pi^{-3/4} \prod_\mu \lambda_{i\mu}^{-1/2} e^{-\frac{1}{2} (r_\mu / \lambda_{i\mu})^2},
\]

where we have introduced the harmonic oscillator length for species \( i \) along the direction \( \hat{r}_\mu \)

\[
\lambda_{i\mu} = \left[ \frac{\hbar^2 |\omega_i - \omega_L|}{m_i d_i^2 u_\mu} \right]^{1/4},
\]

\( m_i \) being the atomic mass of species \( i \). With these wavefunctions, we can compute the collision coefficients via

\[
g_{ij} = \frac{2 \pi \hbar a_{ij}}{m_i m_j} \int \frac{d^3 r}{(2\pi \hbar)^3} \frac{|\phi_i(r)|^2 |\phi_j(r)|^2}{\sqrt{\lambda_{i\mu}^2 + \lambda_{j\mu}^2}}
\]

where \( a_{ij} \) is the scattering length for collisions between species \( i \) and \( j \), and \( \mu_{ij} = m_i m_j / (m_i + m_j) \) is the effective mass.

![FIG. 2. Relative density parameter in an optical lattice for $^{23}$Na, $^{41}$K, and $^{87}$Rb as a function of the lattice frequency $\omega_L$. The relative density parameter is in units of $[m_i e^2 a_0^2/c R_\infty]^{3/4}$ and the laser frequency is in units of $c R_\infty$.](image-url)
Because the harmonic oscillator length (5) for a given atomic species tends to zero when approaching the atomic resonance frequency, tuning the lattice frequency close to the resonance frequency of one atomic species can act as a very strong ‘handle’ with which to tune the relative atomic densities, and hence the collision parameters of the system. The local density per atom, defined as \(1/V_i = \prod_\mu \lambda_{i\mu}^{-1}\), is given by \(1/V_i = \rho_i \hbar^{-3/2} \prod_\mu u_{i\mu}^{1/2}\), where we have introduced the relative density parameter

\[
\rho_i = \left[ \frac{m_i d_i}{\omega_L - \omega_i} \right]^{3/4}.
\]

From the equation for \(1/V_i\), we see that the relative densities of the different atomic species will be independent of the lattice parameters \(u_{i\mu}\), provided only that the wells are sufficiently deep that the Gaussian approximation holds for the lowest Wannier state—generally required for the lattice depth to be large compared to the atomic recoil energy. In Figure 2, we plot the dimensionless density parameter \(\rho_i\) as a function of the lattice frequency across the atomic resonance frequencies. The atomic parameters in this calculation are: \(m_i = \{22.99, 40.96, 86.91\} \times m_p, \; \omega_i = \{0.9720, 0.7472, 0.7351\} \times c R_\infty, \;\) and \(d_i = \{0.9036, 0.8227, 0.8023\} \times e a_0\), respectively, where \(m_p\) is the proton mass, \(e\) the electron charge, \(a_0\) the Bohr radius, \(c\) the speed of light, and \(R_\infty\) the Rydberg constant.

In analogy with previous studies of Bose-Hubbard Hamiltonians [2,12,18], it is safe to assume that for a deep enough lattice and for strictly positive scattering lengths, the ground state of the two-species Bose-Hubbard model having \(N\) atoms per species and \(N\) lattice sites will be an insulator state, i.e. the atoms will be localized in individual wells. The minimum energy arrangement of the two atomic species then depends on the relative strengths of inter- and intra-species collision parameters. In principle, there is a broad variety of possible two-species Mott insulator phases. If, e.g., one species has an extremely weak self-interaction relative to the other interaction parameters, then it may be energetically favorable for all atoms of the first species to occupy a single lattice site with the second species uniformly distributed over the remaining sites. We do not consider such extreme situations and instead focus on the more likely regime where the ground state contains exactly two atoms per site. This regime has two phases: a mixed phase where one atom per species occupies each site, and a separated phase where each site contains two atoms of the same species. We note that the separated phase is unique in that the ground state could be any quantum superposition of different patterns for distributing the two species.

The collisional energy per lattice of the MI state is given by \(E_{s,m} = \frac{1}{N} \langle \psi_{s,m} | \hat{H} | \psi_{s,m} \rangle \right|_{\beta=0}\), where \(N\) is the number of lattice sites. For the case of two lattice sites, the quantum state of the separated MI phase is

\[
|\psi_s\rangle = \frac{1}{2} c_{11}^{\dagger} c_{22}^{\dagger} c_{12}^{\dagger} c_{21}^{\dagger} |0\rangle,
\]

whereas the ground state of the mixed MI phase is

\[
|\psi_m\rangle = c_{11}^{\dagger} c_{12}^{\dagger} c_{21}^{\dagger} c_{22}^{\dagger} |0\rangle.
\]

The energy per lattice site calculated from this two-site state will remain valid as the lattice is scaled up to any even number of sites. The energy per lattice site for the separated-phase is \(E_s = (g_{11} + g_{22})/2\), whereas the energy of a pair of mixed-phase per lattice site is \(E_m = g_{12}\). The condition for a mixed-species ground state is therefore \(E_s/E_m = (g_{11} + g_{22})/2g_{12} > 1\), which can be determined with the help of Eqs. (5) and (6). We see that this condition differs significantly from the free space condition for miscibility [19].

In Figure 3, we plot this ratio as a function of the lattice frequency \(\omega_L\) for the case of \(^{87}\text{Rb}\) and \(^{41}\text{K}\). We use the scattering length data \(a_{\text{Rb-Rb}} = 99 a_0, \; a_{\text{K-K}} = 60 a_0\) and \(a_{\text{Rb-K}} = 163 a_0\), taken from the recent report on a two-species superfluid [10]. In figure 3, the frequency domain is divided into five regions. In region I, both species see a red detuned lattice and the ground state is a separated dual MI. In region II, which spans from \(\omega_L = 0.733663\) to the \(^{87}\text{Rb}\) resonance, both species also see a red detuned lattice but the increase in the rubidium density results in the ground state being the desired mixed phase. This is because the Rb-Rb mean-field energy is proportional to \(\rho_{\text{Rb}}^2\), hence the energy cost of having two \(^{87}\text{Rb}\) atoms can greatly exceed the energy cost of a mixed site despite the much larger \(a_{\text{Rb-K}}\). Region III

![FIG. 3. The ratio of the energies per particle of the separated and mixed phases of a \(^{87}\text{Rb}^{41}\text{K}\) two-species Mott insulator as a function of the lattice frequency \(\omega_L\) taken in units of \(cR_\infty\).](image-url)
lies between the Rb and K resonances, and hence the two species see different potential minima. Here the ground state will be a dual MI, but with the Rb atoms localized at the lattice intensity minima and the K atoms localized at the intensity maxima. In regions IV, which spans from the K resonance frequency to $\omega_L = 0.747631$ and V, both species see a blue detuned lattice, with region IV corresponding to a mixed phase and region V to a separated phase.

The mixed phase occurs only in the vicinity of an atomic resonance: thus, we must make sure that spontaneous emission rates are sufficiently small and remain negligible for experimentally realizable time scales. By averaging the spontaneous emission rate, proportional to the light intensity, over the wavefunction (4), we find with the help of the integral $\int dz \left\{ \frac{\cos^2(kz)}{\sin^2(kz)} \right\} e^{-z^2} = \frac{\sqrt{\pi}}{2} \left( 1 + e^{-k^2} \right)$ and Eqs. (2) through (5) that the spontaneous emission rate of an atom in the ground state of a well of a spherically symmetric lattice with depth $a_h \omega_R$, $\omega_R = \hbar \omega_0^2/(m_c e^2)$ being the atomic recoil frequency, is given in the Gaussian approximation by

$$\gamma_i = \frac{3}{2} \frac{\Gamma_i}{|\omega_i - \omega_L|} \left( 1 \pm e^{-1/\sqrt{\pi}} \right), \quad (10)$$

where $\Gamma_i$ is the natural linewidth of species $i$ and the + and − signs give the result for a red-detuned and a blue-detuned lattice, respectively. For $^{87}$Rb, with $\Gamma_{Rb} = 5$MHz, we find that the average decay rate just to the right of the I-II boundary is $\gamma_{Rb} \approx \alpha(1 + e^{-1/\sqrt{\pi}}) \times 7 \times 10^{-3}$Hz, which, for $\alpha = 20$ gives a lifetime per atom of 0.38s. Similarly, on the blue-detuned side, we find the average decay rate for $^{41}$K, with $\Gamma_K = 6$MHz, just to the left of the IV-V boundary is given by $\gamma_K \approx \alpha(1 - e^{-1/\sqrt{\pi}}) \times 7 \times 10^{-3}$Hz. For $\alpha = 20$ this also gives a lifetime per K atom of 0.35s, roughly the same as for Rb. The time scale for adiabatic passage from a SF to a MI state is typically on the order of 10ns, so working in either the red (II) or blue (IV) detuned regions should be feasible without significant spontaneous heating. We note that once the mixed dual-MI state is reached, the lattice depth could be ramped to a large enough value to suppress tunneling, after which the lattice frequency could be moved much farther away from the atomic resonance while preserving the mixed dual-MI state as a metastable state. With this technique the mixed phase lifetime could be increased significantly to facilitate further experimentation.

In conclusion, we have calculated the ground state configuration of a two-species optical lattice for the case where the number of atoms in each species equals the number of lattice sites and shown that varying the lattice frequency leads to a variety of MI phases. In making the transition to a dual-MI state from a dual-SF state, it is likely that each species will undergo the superfluid-insulator transition at a different lattice intensity. While there are no obvious difficulties with reaching the mixed dual-MI state in this manner, dynamical simulation of the Bose-Hubbard model with the exact Wannier states should be performed to ensure that the system can indeed be driven into the true ground state, a task we plan to carry out in a future work.

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Note: During the preparation of this manuscript we became aware of a related manuscript by Damski et al [20] which reaches similar conclusions but concentrates on different aspects of the proposed technique.\[1\] E-mail: mmoor@ca.harvard.edu.

\[1\] E-mail: hsadeghpour@ca.harvard.edu.

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