Solid phases and pairing in a mixture of polar molecules and atoms

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We consider a mixture of hard core bosonic polar molecules, interacting via repulsive dipole-dipole interaction, and one atomic bosonic species. The mixture is confined on a two-dimensional square lattice and, at low enough temperatures, can be described by the two-component Bose-Hubbard model. The latter displays a rich phase diagram including solid, superfluid, supersolid phases. Here we mainly focus on the checkerboard molecular solid, stabilized by the long range dipolar interaction, and study how the presence of atoms affects its robustness both at zero and finite temperature. We find that, due to atom-molecule interaction, solid phases can be stabilized at both, (much) lower strengths of dipolar interaction and higher temperatures, than when no atoms are present. As a byproduct, atoms also order in a solid phase with same melting temperatures as for molecules. Finally, we find that for large enough interaction between atoms and molecules a paired supersolid phase can be stabilized.

Recent years have witnessed spectacular advances in atomic and molecular physics. Ultracold bosonic atoms have been successfully loaded in one-\cite{1}, two-\cite{2} three-dimensional\cite{3} optical lattices with full control over the hamiltonian parameters. The Mott-insulator (MI) to superfluid (SF) transition has been experimentally observed and extensively studied theoretically\cite{4,5}. Moreover, more recently, new experimental efforts towards trapping two component atomic mixtures in optical lattices have been put forward\cite{6,7}. In the presence of a second component, new fascinating phenomena and exotic quantum phases which cannot be accessed with single species gases, become available\cite{11,12}.

Ultracold polar molecules, interacting via long range dipolar interaction, also represent a fertile ground for the study of novel exotic phases of matter. The dipole-dipole interaction can be tuned and shaped via external static and microwave electric fields\cite{13}. Self-assembled dipolar crystal, dipolar Bose-Einstein condensate, supersolids, are among the phases which can be stabilized with cold polar molecules\cite{14,15}. Besides, interesting applications in fundamental physics, e.g. measurement of permanent electron dipole moment, in quantum chemistry and few-body physics, and in quantum information processing have been envisioned\cite{16}.

Observation of many body quantum phases requires high phase space density of molecules in stable ground states. Cooling high density samples of molecules to quantum degeneracy has proved to be challenging due to their complex internal state structure. Only very recently samples of high density polar and non-polar molecules in the rovibronic ground-state and a selected hyperfine state were realized\cite{17,18}. The Jila group has also reported on observations of dipolar collisions and evidence of spatial anisotropy of dipolar interaction in a sample of KRb molecules with non zero induced dipole moment, i.e. aligned by external electric field\cite{19}. The Innsbruck group has produced a high density sample of Cs$_2$ molecules in their rovibronic ground state trapped into an optical lattice\cite{21}. The procedure used in\cite{21} can be generalized to heteronuclear molecules.

In the present work we consider a novel system consisting of a mixture of polar molecules and one atomic, non reactive species. This system is realizable experimentally since residual atoms naturally occur after molecule formation is completed and can be selectively removed\cite{20}. We are interested in studying how the presence of the atomic species affects molecular solid phases stabilized by dipolar interaction\cite{15}. Hence we consider the case where only reactive atoms are removed from the system, and both, molecules and atoms, are in the absolute (including hyperfine) ground state, so that inelastic collisions are forbidden. Examples of non reactive mixtures include (KRb+Rb), (LiCs+Cs), (RbCs+Cs)\cite{21}.

We consider bosonic polar molecules in a static electric field along the $z$-direction which induces a dipole moment $d$ parallel to the field. Molecules interact via dipole-dipole interaction $V^d(R) = \frac{d^2}{6\pi\varepsilon_0 R^3}$. If the system is confined on the $xy$ plane (by e.g. a strong optical lattice potential along the $z$-direction) in order to avoid instabilities which might arise due to the attractive part of the dipolar interaction, the resulting effective 2D potential is isotropic, $V_{\text{eff}}^d \sim \frac{d^2}{2\pi\varepsilon_0}$. An additional 2D optical lattice with spacing $a$ (which we use as unit length) further confines the system. If atoms are present, they interact with molecules via Van der Waals (anisotropic) interaction, i.e. $V_{\text{vdW}}(r) = \frac{d^2}{12\pi\varepsilon_0 a^6}$, which we describe via an effective s-wave scattering length, and via dipole-dipole interaction $V_{\text{at-mol}}^d(r) = \frac{d^2}{2\pi\varepsilon_0}$. Since atoms have small polarizabilities, dipolar interaction between atoms and molecules can be neglected, provided the electric field is small. Indeed, for DC electric fields $E_{DC} \sim$ kV/cm, and molecular (induced) dipole moment $d \sim 1D$, the dipolar interaction at a typical lattice spacing distance $a \sim 500$ nm is $V_{\text{at-mol}}^d(a) \sim 0.1 - 1$ Hz $\hbar$ (to be compared with molecule-molecule dipolar interaction $V^d(a) \sim$ kHz $\hbar$). At low enough temperature, the system is described by the two-component Bose-Hubbard Hamiltonian:

$$ H = -J_b \sum_{\langle i,j \rangle} b_i^+ b_j - J_a \sum_{\langle i,j \rangle} a_i^+ a_j + V \sum_{i<j} \frac{n_i^b n_j^b}{r_{ij}^3} $$
For example, at repulsive atom-molecule interaction, i.e. atoms and molecules, and consider attractive interaction between densities for atoms and molecules. We work at den-
tative difference from the hard-core case was observed. This can be understood from the fact that, in the limit

\[ -U_{ab} \sum_i n_i^a n_i^b + \frac{1}{2} \sum_i U_{aa} n_i^a n_i^a - 1 \]

\[ -\sum_i \mu^a n_i^a - \sum_i \mu^a n_i^a, \]

where \( b_i \) (\( a_i \)) and \( b_i^\dagger \) (\( a_i^\dagger \)) are bosonic operators for molecules (atoms) with \( b_i^\dagger b_i = 0 \) (the hardcore limit is well justified if one starts with low enough molecular density so that there are no doubly occupied sites \([22]\)), and \( n_i^a \) is the occupation number operator; the first and second terms in Eq. 1 describe the standard kinetic energy with hopping rate \( J_{ab} \) for atoms and molecules respectively, the third term describes the repulsive dipole-dipole in-
teraction between molecules with \( r_{ij} = |i - j| \), the fourth and fifth terms describe the atom-molecule and atom-
atom contact interaction, respectively; \( \mu_{a,b} \) is the chemical potential.

We study model \([11]\) via large scale Monte-Carlo sim-
ulations by the Worm Algorithm \([12,23]\). An Ewald
summation is used in the interaction potential so that no
cutoff in the range of the dipole-dipole interaction is in-
cluded. The complete phase diagram is extremely rich.
It includes mixtures of a molecular solid or supersolid
(SS) with MI or SF of the atomic component, double SF phases, etc. Besides, one expects the presence of paired
phases which have been proved to exist in multicompo-
nent atomic systems (see e.g. \([11,12]\)). In the present
work we mainly focus on the checkerboard (CB) mole-
cular solid phase at density \( n_b = 0.5 \) \([15]\). Our results can
be summarized as follows. Due to atom-molecule in-
teraction, the CB molecular solid appears at weaker dipol-
lar interaction compared to the purely molecular sample.
Correspondingly, solid melting temperatures are higher
than in the absence of atoms. As a byproduct, atoms are
also ordered in a CB solid phase with same melting tem-
peratures as for molecules. We also find that, for large
enough interaction between atoms and molecules a paired
supersolid (PSS) phase, i.e. a SS phase of a composite
(atom+molecule) object, can be stabilized upon doping
of the solid.

Let us first discuss the parameter space we focus on. In
the following we consider \( \mu_a = \mu_b = \mu \) (although the
results discussed below hold in general for \( \mu_a \sim \mu_b \)).
This can be easily achieved by making use of two indepen-
dent lattices for atoms and molecules. We work at den-
sities \( n_a = n_b \sim 0.5 \), in the limit of hard-core molecules
and atoms, and consider attractive interaction between
atoms and molecules, i.e. \( U_{ab} > 0 \). Results still hold for
repulsive atom-molecule interaction, i.e. \( U_{ab} < 0 \), pro-
vided one replaces atoms with holes (and viceversa) in
the following discussion. The hard-core limit is naturally
reached for molecules \([22]\), as we have mentioned above.
For atoms, it can easily be reached via Feshbach reso-
nances. The qualitative results found here, though, are
still valid for soft core atoms as long as \( U_{aa} > |U_{ab}| \gg J \).
For example, at \( U_{aa} \sim 2.5U_{ab} \), and \( U_{ab} = 8J \) no qual-
itative difference from the hard-core case was observed.
This can be understood from the fact that, in the limit

\[ U_{ab} \sim |U_{ab}| \gg J, \]
chemical potentials and already at relatively low atom-molecule attraction, implying that $n_a \sim n_b \sim 0.5$ will be easily satisfied in some regions of the trap.

Let us now turn to the results. In Fig. 1 and 2 we study how the onset of the CB solid is affected by the presence of atoms, upon varying the interaction between atoms and molecules limiting ourselves to values $U_{ab}/J$ for which atomic density also orders, forced by the molecular solid. In Fig. 4 we plot the CB lobes for $U_{ab}/J = 5$ (down triangles), $U_{ab}/J = 8$ (diamonds), and, for reference, in absence of atoms (circles). (We do not investigate the nature of phases outside the lobe, as establishing the full phase diagram of model [11] is beyond the scope of this work). The boundaries of the lobes are determined by monitoring $n_{ab}(\mu_{ab})$ for system sizes $L=12$, 16, 20, 24, 30 (with extrapolation to infinite size when necessary). With no atoms in the system the solid is stabilized for dipolar interaction strength $V/J \gtrsim 3.6$. In the presence of atoms, instead, the solid already exists for $V/J \gtrsim 2.5$ (1.75) at atom-molecule interaction $U_{ab}/J = 5$ (8). Similarly, Fig. 2 shows CB lobes for $U_{ab}/J = 12$ (squares), $U_{ab}/J = 20$ (up triangles), and in absence of atoms (circles). Here the effect of the atom-molecule interaction is even more dramatic. The solid is stabilized for $V/J \gtrsim 0.5$, a factor of 7 smaller than the case of no atoms present in the system! The renormalization of the tip of the lobe towards smaller values of $V/J$ can be understood by noticing that, for $U_{ab}/J$ values considered here, atoms also ordered in a CB solid. One can therefore visualize a solid of a composite object, constituent of one atom and one molecule, which has an effective hopping $J_{\text{eff}} \propto \frac{J_{ab}^2}{J} < J$ responsible of rescaling the onset of CB solid towards smaller dipolar interaction strengths.

The results summarized in Fig.s 1 and 2 have important consequences for experiments, as they show that in presence of non reactive atoms, smaller induced dipole moment is needed in order to stabilize the solid. If one considers (RbK+Rb) mixtures loaded in lattices of depth $V_a \sim 8E_a^s$ and $V_b \sim 7E_b^s$ ($E_a^s$ is the recoil energy of species $a$) and with spacing $a \sim 500\text{nm}$, one has $J_a \sim J_b \sim 80\text{Hz} h$ and $U_{ab}/J \sim 8$, under the assumption $-a_s^\text{Krb-Rb} \sim 150a_0$. In absence of atoms, observation of CB solid requires dipolar interaction $V \gtrsim 3.6J \sim 288\text{Hz} h$ which can barely be reached with fully polarized molecules. In the presence of atoms, instead, one needs $V \gtrsim 1.5J \sim 120\text{Hz} h$, which requires an induced dipole moment $d \gtrsim 0.35D$ (easily achievable in the laboratory with alkali dimers).

The enhanced robustness of the CB solid further manifests itself in larger melting temperatures compared to the case of no atoms present. In Fig. 3 we plot the critical temperature for the melting of the CB solid $T_c/J$ as a function of $V/J$ for $U_{ab}/J = 5$ (diamonds), 8 (down triangles), 12 (squares), 20 (up triangles), and with no atoms (circles). We find that critical temperatures have already saturated at $U_{ab}/J = 8$ and can be up to a factor of 2 larger than in absence of atoms. (We have not resolved $T_c/J$ close to the tip of the lobes where simulations are more challenging.) Within our statistical errors, melting temperatures for the atomic density wave are the same as for molecules, suggesting that a solid of a composite (atom+molecule) object survives up to the melting transition line. To appreciate the effect of atoms let’s consider an induced dipole moment of 0.5 D in a (RbCs+Cs) mixture loaded in a lattice with spacing $a = 400 \text{ nm}$ and depths $V_a(b) \sim 9(7)E_r^a(b)$. With $-a_s \sim 150a_0$ we obtain $J_a \sim J_b \sim 70\text{Hz} h$, $U_{ab}/J \sim 12$, and a solid melting temperature 2 times larger than in absence of Cs atoms.

Next we turn to the discussion of paired phases. Here, we would like to focus on the possibility of realizing a paired supersolid (PSS), i.e. a SS of a composite object formed by an atom and a polar molecule (we remind the reader that a SS is a state of matter featuring both superfluid and crystalline order). In order to check for the existence of a PSS we have calculated the structure factor and the winding number statistics, upon doping the CB solid with interstials, i.e. we work at densities $n_a \sim n_b < 0.5$. The structure factor $S(\mathbf{k})$, which characterizes solid phases, is defined as:

$$S_a(\mathbf{k}) = \sum_{\mathbf{r},\mathbf{r}'} \exp[i \mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')] \langle n_a(\mathbf{r})n_a(\mathbf{r}') \rangle / N_a \tag{2}$$

with $\alpha = a, b$, $\mathbf{k} = (\pi, \pi)$ the reciprocal lattice vector for the CB solid, and $N_a$ the total number of particles of species $a$. Paired superfluidity (PSF), i.e. superfluidity of the composite object, is instead identified by simultaneous finite statistics of the sum of winding numbers squared $<(W_a + W_b)^2 > \propto n_s^{\text{PSF}}$, where $n_s^{\text{PSF}}$ is the superfluid stiffness, and zero statistics of the difference of winding numbers squared $<(W_a - W_b)^2 >$.

![FIG. 3. (Color online) Checkerboard solid melting temperatures with parameters $n_a = n_b = 0.5$, $U_{ab}/J = 5, 8, 12, 20$ diamonds, down triangles, up triangles, squares respectively and, for reference, in absence of atoms (circles).](image-url)
have found that, in order to observe PSF, one needs interactions $U_{ab}/J \gtrsim 12$. The relatively small effective hopping $J_{\text{eff}} \approx \frac{n_s}{L}$, or equivalently large effective mass, associated with PSF, and the presence of long range interaction renders the observation of a PSS a fairly challenging task. On one hand the superfluid stiffness is indeed suppressed by the enhancement of the effective mass of the composite object, on the other hand $V/J_{\text{eff}} \gg 1$ gives rise to metastability \[15\]. Since we have found that the CB solid remains robust against temperature and doping also close to the tip of the lobe, the optimal situation for the observation of PSS is indeed in this region of the phase diagram, where metastability is much less severe.

In Fig. 4 we plot $S_b(\pi, \pi)$ (right axis) and $n_s^{\text{PSF}}$ (left axis) as a function of density with parameters $J_a = J_b$, $n_a = n_b$, $U_{ab}/J = 12$, $V/J = 0.5$, linear system sizes $L=12$ (circles), 16 (squares), 20 (up triangles), 24 (down triangles), 30 (diamonds), and inverse temperature $\beta = L/J$. There exist a finite range of densities for which both $S_b(\pi, \pi)$ and $< (W_a + W_b)^2 >$ are finite. (We have checked that $S_a(\pi, \pi)$ and $< (W_a - W_b)^2 >$ are simultaneously finite and zero respectively.) While for $S_b(\pi, \pi)$ there are not finite size effects within errorbars and provided $L > 12$, for $n_s^{\text{PSF}}$ finite size effects are present. As $L$ increases so does $n_s^{\text{PSF}}$, signifying that in the thermodynamic limit $n_s^{\text{PSF}}$ is indeed finite. Pronounced size effects, which can be seen even at densities $n_b \sim 0.43$, where the CB solid is disappearing, are most likely due to low Kosterlitz-Thouless (KT) temperatures for the PSF-normal transition. This is further justified by finite temperature results. We have indeed found that at $n_a = n_b \sim 0.45$, $T_{KT} \lesssim 0.05J$ (not shown here).

The large effective mass of the composite object enhance metastability. As a result it is computationally challenging to establish how far the PSS phase extends. We have found the presence of many metastable phases already at $V/J \sim 1$ and $n_b \lesssim 0.5$, not very far from the tip. These states are characterized by the presence of various grain boundaries and/or extended defects. These observations are also relevant in the context of PSS with polar molecules in bilayered systems with no inter-layer tunneling \[24\]. In \[24\] the authors study soft core bosons with in-plane nearest neighbor repulsion and inter-layer nearest neighbor attraction. By using a combination of mean field Gutzwiller approach and exact diagonalization to study an effective hamiltonian of pairs, they predict the existence of a PSS phase. Polar molecules on bilayers with zero inter-layer tunneling can be interpreted as a two-component mixture with same intra-species long range interactions while the inter-species one, also long range, can be independently tuned (by changing the distance between layers). The model studied here differs from the one describing polar molecules in bilayers in that both, atom-atom and atom-molecule interactions are short range. We expect the metastability to become even more severe when all interactions are long range, as it happens in the case of bilayers, making the existence of a PSS phase questionable and surely extremely challenging to observe both in simulations and experimentally. Preliminary simulations validate this expectation.

In conclusion, we have studied a mixture of hard core bosonic polar molecules and one atomic non reactive bosonic species, confined on a two-dimensional square lattice. We have shown that the presence of atoms renders the CB molecular solid phase more robust both at zero and finite temperature, i.e. CB forms at lower strengths of dipolar interaction and survives at higher temperatures. We have estimated that this effect could be sizeable in current experimental setups. As a byproduct, atoms also order in a solid phase with same melting temperatures as for molecules. We have also established that for large enough interaction between atoms and molecules a PSS can be stabilized. We expect the observation of the latter to be experimentally challenging due to the presence of metastability and the low temperatures at which the superfluid component disappears.

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