Atomic and molecular matter fields in periodic potentials.

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This paper deals with the conversion between atoms and molecules in optical lattices. We show that in the absence of collisional interaction, the atomic and molecular components in different lattice wells combine into states with macroscopic condensate fractions, which can be observed as a strong diffraction signal, if the particles are abruptly released from the lattice. The condensate population, and the diffraction signal are governed not only by the mean number of atoms or molecules in each well, but by the precise amplitudes on state vector components with different numbers of particles. We discuss ways to control these amplitudes and to maximize the condensate fraction in the molecular formation process.

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INTRODUCTION

The immersion of degenerate gases in laser induced periodic potentials has led to numerous beautiful studies of both many-body and few-body phenomena, including squeezing, the superfluid-insulator phase transition, and collisionally induced collapses and revivals of the phase of a quantum field with few quanta (atoms) [1, 2, 3]. We have previously shown [4] that with an average population of approximately a single atom per site or less, the removal of atom pairs from the lattice may be used to reestablish a robust atomic interference pattern even long times after dephasing has taken place due to collisional interaction among the atoms. This works, because the remaining zero and one-atom components of the quantum state have experienced no interaction, and hence no dephasing. In the same work we showed that if this removal occurs by formation of a bound molecular state, a molecular condensate may be produced and detected when the lattice is removed. The incommensurate Rabi frequencies governing the dynamics among different population components present a major obstacle towards extending the results to the case with more particles in every lattice well, and the restriction to superposition states dominated by a vacuum component and a single atom and a single molecule component on every lattice site was crucial for the analysis in [4].

In the present paper we first motivate our work by a brief discussion of the recent experimental and theoretical work on formation of molecules in cold atomic gases. We then summarize the analysis of the connection between the quantum states of particles in individual wells of a periodic potential, diffraction patterns, and the macroscopic population of a single condensate wave function. Finally, we turn to the process of converting more than a single pair of atoms into molecules in every lattice site, and we present a method to do this with maximal possible coherence of the final state. We shall not make use of atomic interactions and of the Mott-insulator phase transition dynamics, which can prepare an exact integer number of atoms per site and hence also an exact number of molecules [5, 6]. We assume instead that the lattice potential is applied to the condensate too fast for the Mott insulator transition to happen, but slowly enough that the spatial motion of the individual atoms and molecules follow the lowest energy state of the potential.

CONVERSION BETWEEN TRAPPED ATOMS AND MOLECULES

By carefully designed photoassociation [7], which makes use of the fact that the two-atom component is in a fully determined initial state, it is possible to convert atoms in a condensate effectivelly into molecules. By tuning a magnetic field across an atomic Feshbach scattering resonance in $^{85}$Rb, Donley et al. [8] observed a significant production of energetic atoms, and a loss of atoms, which could be well accounted for by the production of a loosely bound molecular system [9, 10, 11]. Also thermal and condensed $^{133}$Cs atoms have been converted into molecules by tuning of a magnetic field, and the fermionic atoms $^{40}$K and $^6$Li have been converted into bosonic diatomic molecules [12, 13]. The long lifetimes of molecular states formed by fermionic atoms [14] suggest unique possibilities to study condensation of the molecular component and to convert the system back to the atomic system in the BCS superfluid state [15].

All of these above mentioned experiments and theories deal with large quasi-homogeneous systems, for which mean field theories are well justified, and for which the atoms and molecules, ideally, populate only a single spatial quantum state. It has been proposed to study the molecular formation processes after the atoms have been subject to a periodic potential in form of an optical lattice [16, 17]. The clustering of the atoms in the
potential wells increases significantly the two-atom density correlations of the system and hence the molecular formation rate. In addition, the quantization of the motion in the narrow wells, makes the photoassociation process a transition between well-resolved discrete states with resulting better possibilities to control the coherence properties, and avoid ‘rogue’ photoassociation. From an experimental point of view our molecules can be prepared in a deeper bound molecular state, selected coherently by the coupling laser fields. In [13] it is proposed to invoke the Mott-insulator dynamics and prepare a system with precisely two atoms per well, to turn these atoms into a single molecule, and to subsequently melt the molecular Mott insulator and form a molecular condensate. In [4, 18], we do not apply the Mott dynamics, but we rather accept the fluctuating atom number in each well, and use the fluctuations as a seed to prepare mean molecular fields in every single well, which combine to a macroscopic condensate upon release of the atoms. Cold atoms in lattices offer a number of interesting possibilities for many-body physics, quantum optics and quantum information studies, and we believe that the conversion to molecules will provide extra possibilities to study multi-component dynamics, and, e.g., mixed Bose-Fermi systems, and for detection of low-temperature phase transition dynamics.

DIFFRACTION AND CONDENSATE FRACTION IN A LATTICE SYSTEM

Apart from the restriction of the atomic spatial dynamics induced by the confining potential, the periodicity is the most characteristic feature of the optical lattice, and in this section we shall summarize some simple properties related to this periodicity.

Splitting of number states and coherent states

We assume that we have initially a quasi-homogeneous Bose-Einstein condensate at rest. This condensate may contain a definite number $N$ of atoms, which fragments with a probability distribution $p(n)$ for the occupancy $n$ of each of a large number of wells. The state of the system is described by a quantum mechanical many-body wave function, and the contents of each well is necessarily entangled with the contents of the other wells. If, instead the entire many body wavefunction is described as a coherent state, i.e., an eigenstate of the atomic annihilation operators $\hat{\Psi}(x)$, in the absence of interaction among the atoms, this state factors exactly into a product state of coherent states populating each well. Each site is in that case populated by a superposition \( \sum_n c_n |n\rangle \) of zero, one, or more atoms, where the amplitudes $c_n$ can be parametrized by a single coefficient $\beta$: $c_n = \exp(-|\beta|^2/2)\beta^n/\sqrt{n!}$. As long as the lattice potential is kept deep enough, the factorization of the total system into the contents of each well is maintained in the future evolution of the system, and hence we need only determine the state vector evolution for the combined atom-molecule system in each well, and finally obtain the macroscopic properties from the resulting product state.

The macroscopic coherent state is a quantum mechanical superposition of states with a total atom number $N$ distributed according to a Poisson distribution, and with a specific relative phase $\phi$ between different number state components. Such a superposition state does not have a definite total number of atoms, but the number fluctuations are insignificant in comparison with the total number $N$ in the limit of large condensates, and one will not see any difference between predictions based on the coherent state and on a state with precisely $N$ atoms. Remarkably, this argument also applies when the condensate is being split over many wells, where the relative fluctuations are not insignificant. The reason is that the poissonian fluctuations in atom number on each lattice site account properly for the entanglement of the content of the site with the remaining system, and the relative phase between number states assumed by the coherent state Ansatz leads to correct predictions for the interference between different wells, even though this interference is due to entanglement rather than mean-fields [20, 21]. A more formal argument states that a Poissonian mixture of number states is identical to a mixture of coherent states with random phase and the same mean population, and hence predictions for the outcome of any time evolution and subsequent measurement on states picked at random from either ensemble will be strictly identical [21], also when this evolution involves splitting the system in smaller components. We shall apply the coherent state Ansatz throughout the paper.

Atomic and molecular field operators in position, momentum and Wannier state representation

When the optical lattice is applied slowly to the zero momentum condensate, it evolves into the the zero momentum state of the lowest Bloch band of the potential, which is, in case of a deep potential modulation, a real superposition of Wannier states localized in each well, $\phi_m(x) = \phi_0(x - mL)$.

If the atoms occupy only the ground states of the wells of the lattice, it is convenient to introduce the discrete set of atomic field operators, $a_m$, that remove atoms from the Wannier mode functions rather than $\hat{\Psi}(x)$, which remove atoms from specific locations in space,

$$a_m = \int \phi_m(x)\hat{\Psi}(x)dx. \tag{1}$$

The mode operators obey the standard commutator re-
lations \([a_m, a_m^\dagger] = \delta_{m,m'}\).

Let \(|n\rangle_m\) denote the state with \(n\) atoms populating the spatial wavefunction \(\phi_m(x)\). In the state \(\sum_n c_n |n\rangle_m\), the mean value of the mode annihilation operator in the well equals \(\alpha := \langle a_m \rangle = \sum_{n=1}^\infty c_n^* c_n \sqrt{\bar{n}}\), the mean number of atoms equals \(\bar{n} := \langle a_m^\dagger a_m \rangle = \sum_{n=1}^\infty |c_n|^2 n\), and in the product state \(\Psi = \Pi_m (\sum_n c_n |n\rangle_m)\) we have the interwell coherence \(\langle a_m^\dagger a_m \rangle = \alpha^* \alpha\) for \(m \neq m'\).

The condensate population is defined as the largest eigenvalue of the one-body density matrix \(\langle \hat{\Psi}^\dagger(x)\hat{\Psi}(x') \rangle\), and in the basis of localized Wannier states, the mean occupancy \(\bar{n}\) of the wells appears in the diagonal and the squared mean field amplitude \(|\alpha|^2\) appears in all other positions in this matrix. In a system with \(N_L\) wells, the eigenvector with equal amplitude on each Wannier function has the largest eigenvalue of \((N_L - 1)|\alpha|^2 + \bar{n}\), i.e., in the limit of large \(N_L\) the condensate fraction is \(|\alpha|^2 / n\), in agreement with the value predicted by the off-diagonal long-range order, \(\langle a_m^\dagger a_m \rangle = |\alpha|^2 = \bar{n} \cdot (|\alpha|^2 / \bar{n})\). This analysis of condensate population and fraction also applies to the molecules if \(\bar{n}\) and \(\alpha\) are replaced by the molecular mean population \(\bar{n}_M\) and the molecular mean field \(\alpha_M\). Note that if the potential is realized slowly, the many fragments will combine together to form a single condensate with the population just identified.

In [4], we introduce \(\hat{\Psi}(k) = \frac{1}{\sqrt{2\pi}} \int dx e^{ikx} \hat{\Psi}(x)\), and we present a derivation of the momentum distribution of the particles after release from the periodic potential,

\[
\langle \hat{\Psi}^\dagger(k) \hat{\Psi}(k) \rangle = N_L |\phi(k)|^2 \left( \bar{n} - |\alpha|^2 + |\alpha|^2 \sum_q \delta(k - q\frac{2\pi}{L}) \right),
\]

where \(\phi(k)\) is the Fourier transform of the Wannier function \(\phi_0(x)\). The sum over integers \(q\) gives rise to a comb at lattice momenta \(\frac{2\pi}{L}\) with a modulation proportional to the square of the mean field amplitude \(|\alpha|^2\). This comb sits on top of a flat background caused by the incoherent population \(\bar{n} - |\alpha|^2\) of the wells, and the whole distribution is comprised within the width of the single well momentum distribution \(|\phi(k)|^2\).

Using a lattice for the study of this system is not only convenient for the tailoring of the association process, or for the possibility to observe Mott insulator dynamics: the multi-well fragmentation of the system leads to a diffraction pattern with a clear identification of the coherence properties and the condensate fraction of the system. By counting the released atoms or the molecules on a position sensitive detector the values of \(\bar{n}\) and \(\alpha\) and thus the condensate fraction of both atoms and molecules can be determined.

CONVERTING SEVERAL ATOMS INTO MOLECULES

We now turn our attention to the formation of molecules. The above section provides the necessary connection to the macroscopic signals and condensate properties, and we shall hence treat the dynamics in a single well, only.

Theoretical maximum values

Let us first assess the theoretical maximum number of molecules and maximum mean molecular field, in the case where an atomic state \(\sum_n c_n |n\rangle\) is made subject to the association process. For even \(n\) it is possible to remove the entire atomic component and create \(k = n/2\) molecules, whereas for odd \(n\) one may obtain \(k = (n - 1)/2\) molecules, and one is left with a single atom. We can write the maximum number of molecules for each atomic number state component as \(k = (n/2 - 1/4) + (-1)^n/4\), and thus obtain the maximum expectation value of the number of molecules,

\[
\langle n_M \rangle \leq \frac{n}{2} - \frac{1}{4} + \frac{1}{4}((-1)^n). 
\]

For a coherent state with mean atomic population \(\bar{n}\), we get

\[
\langle n_M \rangle \leq \frac{\bar{n}}{2} - \frac{1}{4} + \frac{1}{4}e^{-\frac{\bar{n}}{4}},
\]

and for a thermal state (exponential distribution) with mean atomic population \(\bar{n}\), we get

\[
\langle n_M \rangle \leq \frac{\bar{n}}{2} - \frac{1}{4} \frac{2\pi}{2\bar{n} + 1}.
\]

The mean molecular field amplitude is maximized if the atoms are maximally converted to the state

\[
|\Psi\rangle = \sum_k r_{2k}|k\ mol\rangle + \sum_k r_{2k+1}|1\ atom, k\ mol\rangle,
\]

and if the amplitudes on the molecular states are all real and positive \(r_n = |c_n|\), so that we obtain \(\alpha_M = \sum_k (r_{2k} - \sqrt{r_{2k}^2 + r_{2k-1}^2 + r_{2k+1}^2})\). We have not obtained a closed analytical form for this maximum, but for a coherent atomic input state we find that the square of the maximal mean molecular field approaches \(|\alpha_M|^2 \sim \langle n_M \rangle - 1/8\) in the limit of large \(\bar{n}\).

Numerical analysis

The above quoted results are theoretical maximum values. The purpose of this section is to investigate how
FIG. 1: Incoherent and coherent molecular components prepared by photoassociation of atoms. The number of molecules (solid line) and the squared norm of the mean molecular field (dashed line) as a function of time are shown, starting at time zero with a coherent atomic state with a mean number of atoms of 3. Time is measured in units of the reciprocal coupling constant $\chi^{-1}$.

close it is possible to arrive to these values in an experiment. We have solved numerically the time evolution of the simple photoassociation Hamiltonian

$$H_{PA} = \chi (a^2 b^\dagger + (a^\dagger)^2 b)$$  \hspace{1cm} (7)

where $a(b)$ is the operator of annihilation of an atom (molecule) at an arbitrary site. The site index is suppressed for convenience, since exactly the same process takes place at every site in the lattice. Starting with a coherent atomic state, the Hamiltonian (7) introduces in every lattice well a superposition state $\sum c_{n,n_M}(t)|n,n_M\rangle$, from which the mean number of molecules and the mean molecular field is readily calculated. Atom-atom interactions and interactions between the atoms and molecules are readily incorporated in these calculations, but they are set to zero in the present study, since our main focus is on processes which are fast on the time scale of interactions.

Fig. 1 shows the results of a calculation with a constant resonant coupling. The initial atomic state is a coherent state with $\pi = 3$ atoms per well. The figure shows that, at first, atoms are effectively converted into molecules, but the conversion stops before the maximum possible value of 1.251 molecules per site is achieved, and the square of the mean field does not reach its maximum which we have computed to be 1.165. The long time dynamics shows a pattern of collapses and revivals of large amplitude oscillations, linked with the different Rabi frequencies between the different number components of the quantum state of the system.

In order to increase the number of molecules, we propose to apply a frequency chirp to the photoassociation lasers across the resonance. If this chirp is slow enough, the system will follow adiabatically the eigenstates of the time dependent Hamiltonian. If the lasers are initially detuned to a too low energy, the photon dressed state with $n$ atoms has lower energy than the state with $n - 2$ atoms and one molecule etc., whereas at the end of the chirp, these are the states with the highest energies, and the lowest energy state is the one with none or a single atom, and $n/2$ or $(n - 1)/2$ molecules. The chirp applies to all atomic $n$-components at once, and if the chirp is slow, the system follows the lowest energy eigenstate throughout the process, and the atoms are all converted into states with the largest possible number of molecules. Fig. 2. shows the energy eigenstates during a chirp, where the coupling strength $\chi(t)$ is turned on and off as a $\sech$-function of time, and the detuning initially takes a value which is minus the maximum coupling Rabi frequency $\chi_{\max}$, and which is scanned linearly in time to a final value equal to the maximum coupling Rabi frequency. The plot shows the states with 4 atoms, 2 atoms and a molecule and 2 molecules, which are coupled by the
lasers. Similar spectra exist for all other initial number
states for the atoms.

The phases are of course determined by our computation,
but it is a non-trivial task to act on the system
and individually control these values. We shall instead
present a strategy, inspired by composite pulse and spin
echo techniques applied in nuclear magnetic resonance
studies \[22\], that accomplishes full phase control of the
system.

\begin{center}
\textbf{Cold atoms} \rightarrow 'hot' \textbf{molecules} \rightarrow 'hot' \textbf{atoms} \rightarrow
'cold' \textbf{molecules}
\end{center}

Looking at Fig. 2., we note that the unknown phases
are simply given by the areas between the horizontal line
and the time dependent energy levels. Observing that
the time dependent energy spectra in the figure are sym-
metric with respect to inversion in the crossing of the
dashed diabatic lines, we can thus identify a means to
cancel these phases: After one chirp, the purely atomic
components have followed the states with the lowest
energy to become components with zero or one atom and a
maximum number of molecules. If one repeats the chirp,
on the system, which now starts with molecules, i.e., the
upper state on the left, the system will now follow the
highest energy state and return to the purely atomic com-
ponent, which is the highest state in the right part of
the figure. The accumulated phase in the second chirp will
exactly annihilate the one of the first chirp, and since
this is true for all initial atomic number states, the com-
bination of the two chirped interactions do not change the
state of the system. Now, let us instead perform the sec-
ond chirp at half the rate of the first one. In this case,
the phases accumulated on all components are twice the
negative of the phases of the first chirp. The atoms have
been recreated, but they have picked up phases so that
the atomic system is actually not coherent any more, and
if it is released at this point, its condensate fraction will
be poor. We are not finished yet, however, and perform-
ing a third chirp on the system at the original chirp rate,
we again turn as many atoms as possible into molecules,
and the phases of the two rapid chirps and the single slow
chirp will cancel to produce a real linear combination of
molecular states.

\begin{figure}
\includegraphics[width=\textwidth]{fig3}
\caption{Number of molecules (solid line) and square of mean
molecular field (dashed line) as functions of time during adia-
batic chirp across the photo association resonance. The initial
atomic state is a coherent state with a mean population of
three atoms per well. The molecular population at the end of
the chirp is within reach of the theoretical maximum, but the
mean field is clearly much smaller than we could have hoped
for.}
\end{figure}

An example of the results of this process is shown in
Fig. 3, where we observe that more molecules are pro-
duced than in the case of constant couplings shown in
Fig. 1. Time is given in units of the maximum value of
the time-dependent coupling coefficient $\chi_{\text{max}}^{-1}$.

The chirp may be carried out in different ways, and we
do not claim to have identified the optimum. The figure
shows that we produce close to the maximally expected
number of molecules but it also shows, that the coherent
component is very weak. As discussed in \[2\], the short
range interaction between atoms causes a phase evolution
of the $c_n$ amplitudes with a frequency $\frac{U}{2}\sqrt{n(n-1)}$, leading
to a reduction of the atomic field amplitude $\alpha$ since the
terms $c_{n-1}^*c_n\sqrt{n}$ acquire different complex phases. The
resulting disappearance of atomic interference has been
observed in experiments \[1\], and subsequent revivals
when the phase differences reach multiples of $2\pi$ have also
been shown \[3\]. In our case, the loss of mean field is a
consequence of the different complex phases attained by
the different state vector amplitudes as a result of the
association process. The adiabatic process, following the
lower solid curve in Fig. 2., implies that a phase factor
proportional to the area between this curve and the hori-
zonal line will appear on the molecular state. This area,
end hence the phase, will be different for the adiabatic
states obtained with other number state components. If
the lattice is removed at the end of the process depicted
in Fig. 3., the molecules will populate a number of differ-
ent spatial states, and the zero momentum macroscopic
state will only account for about 1/8 of the molecules.
FIG. 4: Number of molecules (solid line) and square of mean molecular field (dashed line) as functions of time during three adiabatic chirps across the photo association resonance. The initial atomic state is a coherent state with a mean population of three atoms per well. The first chirp, of duration $t = 200\chi_{\text{max}}^{-1}$, is as depicted in Fig. 3. The second chirp with duration $t = 400\chi_{\text{max}}^{-1}$ converts the molecules back into atoms, and the last chirp produces the molecules again, but now with a sizable mean field, as shown by the dashed line.

FIG. 5: Same as Figure 4., but with 5 times faster chirps. The outcome is not as good as in Figure 4, but still better than the results of a constant coupling, shown in Fig. 2.

chirp schemes, e.g., as function of the total duration available for an experiment.

DISCUSSION

In summary, we have presented a method to prepare a molecular system in a macroscopically populated quantum state. The molecular condensate exists due to the simultaneous existence on every lattice site of different number state components, which are inherited from the number fluctuations in the atomic system. The system is too small to be treated by a mean field Ansatz, and a separate derivation of the mean value of the field operator indeed shows that the phase is ill-defined. Due to the deterministic character of the problem - there is no decoherence or loss at play - we were able to apply a controlled interaction, in which molecules were first created, then removed, and finally created again with a well-behaved mean field. This technique, and possible rapid versions which utilize composite pulse sequences, may be interesting to study in their own right for this system, and we believe that they may also be highly relevant in other studies with optical lattices with small occupation numbers on each site, e.g., in connection with simulations of quantum magnetism and superfluidity.

Note added -. After submission of this paper, the first experimental results were reported on observation of molecular condensates, formed by association of fermionic potassium and lithium atoms [23].

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