Atoms and molecules in lattices: condensates built on a shared vacuum

Tilman Esslinger and Klaus Mølmer

1. Institute for Quantum Electronics, ETH Zurich
CH-8093 Zurich, Switzerland
2. QUANTOP, Department of Physics and Astronomy,
University of Aarhus
DK-8000 Århus C, Denmark

In optical lattices where each site is occupied in its lowest energy state by a superposition of zero, one and two atoms, one can in a controllable manner convert the atomic pair into a molecule while retaining the vacuum and one-atom amplitudes. The microscopic quantum coherence on each site between the vacuum and the single molecule component leads to a macroscopically populated molecular condensate when the lattice is removed.

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In this paper we present a simple quantum optical analysis of the diffraction pattern of atoms and molecules released from a periodic potential. We show that removal of atom pairs from a lattice with low filling fraction may be used to reestablish a robust atomic interference pattern after dephasing has taken place due to collisional interaction. If the removal occurs by photoassociation, a molecular condensate may be produced and detected when the lattice is removed. We do not make use of the Mott-insulator phase transition dynamics \[1, 2\]. On the contrary, the existence of a superposition of vacuum and one or several atoms or molecules on every lattice site is crucial, and our proposal works ideally in the absence of atomic interactions.

Assume that an atomic Bose-Einstein condensate has been exposed to a lattice potential, which was turned on slowly enough for the spatial motion of the atoms to remain in the lowest energy state but too fast for the Mott insulator transition to have happened. This system is described as a single condensate in the \( q = 0 \) state of the lowest Bloch band of the potential, which is, in case of a deep potential modulation, a superposition of Wannier states localized in each well. We assume that all the atoms in the \( m^{th} \) well are well described by the \( m^{th} \) Wannier function \( \phi_m(x) = \phi_0(x - mL) \).

If the entire body wavefunction is described as a coherent state, i.e., an eigenstate of the atomic annihilation operators \( \hat{\Psi}(x) \), the state remains a coherent state, which factors exactly into a product state of coherent states populating the Wannier states, i.e., each site is populated by a superposition \( \sum_n c_n |n\rangle_m \) of zero, one, or more atoms, where the amplitudes \( c_n \) can be parametrized by a single coefficient \( \beta \): \( c_n = \exp(-\beta^2/2)\beta^n/\sqrt{n!} \). Such superposition states do not reflect the conservation of atom number. The fluctuations in atom number on each lattice site, however, reflects the entanglement of the wells due to the splitting of the atoms - and it is a remarkable feature of coherent states that this entanglement is well accounted for by a product state (because a formal projection on a total number eigenstate with very many atoms, which indeed produces a non-separable state, does not change our physical predictions).

Since atoms are bound to occupy the ground states of the wells of the lattice, it is convenient to introduce the discrete set of atomic field operators that removes atoms from the Wannier mode functions rather than from a specific location in space. These are defined as

\[
a_m = \int \phi_m(x)\hat{\Psi}(x)dx, \tag{1}
\]

and they obey the standard commutator relations \([a_m, a_n^\dagger] = \delta_{m,n'}\).

With the product wave function \( |\Psi\rangle = \Pi_m (\sum_n c_n |n\rangle_m) \), the mean value of the mode annihilation operator in each well equals \( \alpha := \langle a_m \rangle = \sum_{n=1}^{\infty} c_n^* c_n \sqrt{n} \), the number of atoms in each well equals \( n := \langle a_m^\dagger a_m \rangle = \sum_{n=1}^{\infty} |c_n|^2 n \), and we have the inter-well coherence \( \langle a_m^\dagger a_m \rangle = \alpha^* \alpha \) for \( m \neq m' \). The short range interaction between atoms is a weak perturbation which causes a phase evolution of the \( c_n \) amplitudes with a frequency \( \frac{\hbar}{2} n (n-1) \). In the absence of tunneling between wells, the product form of the wave function remains exact, but the phase evolution leads 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 \[3, 4\], and subsequent revivals when the phase differences reach multiples of \( 2\pi \) have also been shown \[3\].

We now turn to the special case of on the order of or less than unit mean occupancy of each well. The amplitudes on states with three or more atoms are small and we shall mainly focus on the lower \( n \) components. Since \( |c_0 c_1 + c_1^* c_2 \sqrt{2}| \) may be smaller than \( |c_0^* c_1| \), it seems an interesting experiment to remove the component with two atoms from the state vector of each well. This can be done by a carefully designed photoassociation process \[2\] \[1\], making use of the fact that the two-atom component is in a fully determined initial state. Neglecting
higher atomic occupancies, the resulting state vector becomes a product of two-species superposition states:

$$|\Psi\rangle = \Pi_m(c_0|\text{vacuum}\rangle + c_1|1\text{ atom}\rangle + c_2|1\text{ molecule}\rangle)_m,$$

(2)

In the state (2) we can readily determine the mean atomic amplitude and population, $\alpha = c_0^*c_1$ and $n = |c_1|^2$. The mean amplitude acquires a finite robust value, which is not corrupted by the atom-atom interaction before or after the photoassociation. We may thus wait arbitrarily long before we remove the atomic pairs, and still the coherence reappears. Technically, we define condensate population as the largest eigenvalue of the one-body density matrix, and applying the basis of Wannier states, this matrix has the mean occupancy of the wells $n$ in the diagonal, and the squared amplitude $|\alpha|^2$ in all other positions. In a system with $N$ wells, the eigenvector with equal amplitude on each Wannier function has the largest value of $(N-1)|\alpha|^2 + n$, i.e., in the limit of large $N$ the condensate fraction is $|\alpha|^2/n$. This is also the value predicted by the off-diagonal long-range order, $\langle a_N a_m^\dagger \rangle = |\alpha|^2 = n \cdot (|\alpha|^2/n)$. It is interesting to note that in cases where $c_2$ is sufficiently dephased, photoassociation increases the value of $|\alpha|^2$ and hence both the total occupancy of the condensate and the condensate fraction increase by the removal of atoms. The state before photoassociation is a perfect, pure quantum state of the system, and it is related to the perfect condensate state by a unitary process - but the system cannot be described by a Gross-Pitaevskii wave function, and the one-body density matrix cannot distinguish a deterministically evolved phase from, e.g., a finite temperature decoherence effect.

Let us now turn our attention to the molecules prepared in the lattice. It is clear from the state (2), that the atomic and molecular states are quite equivalent: there is a molecular population $n_M = |c_2|^2$ per lattice site, and there is a mean molecular field of $\alpha_M = c_0^*c_2$. The above analysis of condensate population and fraction thus applies to the molecules if $n$ and $\alpha$ are replaced by $n_M$ and $\alpha_M$. The preparation of molecular condensates has received quite some interest, in particular with the recent experiments at JILA, where coherent oscillations between an atomic condensate and a condensate of very loosely bound molecules are observed and agree well with a detailed theory of collision dynamics near a Feshbach resonance, see also. In addition to the detailed binary collision dynamics, it is necessary to represent correctly the many-body dynamics in the case where many atoms are converted into molecules. In comparison, our analysis is much simpler since there is either nothing or just a single atom or just a single molecule in every lattice site (as long as the truncation of the atomic state above two atoms is a valid approximation). There is accordingly just a single population independent coupling between the relevant states $|n = 2\text{ atoms}\rangle$ and $|n_M = 1\text{ molecule}\rangle$. From an experimental point of view our molecular states can be prepared in a deeper bound molecular state, selected coherently by the coupling laser fields rather than by Feshbach collision dynamics, and they are prepared in a spatial condensate for which the phase coherence can be readily detected by diffraction experiments similar to the ones applied to atoms in (3), see below.

Our proposal may also be compared with the suggestion by Jaksch et al., where the Mott-insulator dynamics is used to prepare a lattice with precisely two atoms per site, these atoms are transferred into a molecule, and the molecular Mott-insulator is subsequently melted to yield the molecular condensate. Our proposal emphasizes a simple but spectacular phenomenon: a zero-quantum amplitude is enough to yield a mean field, and hence the condensate establishes itself simply by the production of molecules out of the two-atom component of the atomic state. This is so, because the atomic vacuum is of course also a molecular vacuum (as well as a vacuum for larger molecules, butterflies and freight trains). On the experimental side, the molecular condensate may ideally be created from an atomic condensate of non-interacting particles, and we only need to transfer the atoms slowly enough into the lattice to have a well-specified initial state for the photo-association process.

The analysis is quite simple and fully analytical in the limit of very weakly populated sites. Also for a larger mean number of atoms on each site the conversion into molecules will lead to a molecular mean field, i.e., a molecular condensate. To investigate this, we have solved numerically the time evolution of the simple photoassociation Hamiltonian

$$H_{PA} = \sum_m \chi(a_m b_m^\dagger + (a_m^\dagger)^2 b_m)$$

(3)

where $b_m$ is the operator of annihilation of a molecule at site $m$. Starting with a coherent atomic state, the Hamiltonian (4) 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. The results of such calculation are shown in Fig.1. It shows the number of molecules and the absolute square of the molecular field as functions of time: the conversion is oscillatory in time, and to optimize the coherent molecular component, one should stop the photoassociation process after a time interval depending on the number of atoms per lattice site. For simplicity we did not incorporate atom-atom interactions or interactions between the atoms and molecules in these calculations since our main focus is on lattices with low population.

In the above analysis we have shown that the on-site mean field amplitude and population mathematically determine the condensate fraction. These quantities are directly measurable by the diffraction occurring when the
particles are released from the lattice. The detection of the diffraction pattern has precisely been the hallmark of coherence \[\Psi\] and coherence decay and revivals \[\hat{\Psi}\] in experiments, and we present here a brief theoretical description, linking the diffraction pattern directly to our quantities \(n\) and \(\alpha\).

If one introduces a complete and orthonormal set of wavefunctions \(\{\phi_{m,l}\}\), where \(\phi_{m,0} = \phi_m\) are the Wannier functions applied above, and \(\{\phi_{m,l}\}\) with \(l > 0\) are higher excited states centered on the \(m^{th}\) lattice site, the commutator relations \([\hat{\Psi}(x), \hat{\Psi}^\dagger(x')] = \delta(x-x')\), lead to \([a_{m,l}, a_{m',l'}^\dagger] = \delta_{m,m'} \delta_{l,l'}\), with the mode annihilation operators defined through

\[
a_{m,l} = \int \phi_{m,l}(x) \hat{\Psi}(x) dx.
\]

We shall need expressions for the position dependent annihilation operators in terms of the mode functions,

\[
\hat{\Psi}(x) = \sum_{m,l} a_{m,l} \phi_{m,l}(x) = a_m \phi_m(x) + \delta \hat{\Psi}(x),
\]

where in the second line we include only explicitly the Wannier state \(\phi_m\), where \(m\) enumerates the well which is most close to the position coordinate \(x\), and where the ‘noise-term’ \(\delta \hat{\Psi}(x)\) represents all other modes, which are either unoccupied (\(l > 0\)) or which have no support at the location \(x\), \((m \neq m)\). This noise term does not contribute to atom counting signals.

In experiments on trapped condensates, the spatial properties of the atomic cloud are detected by releasing the cloud and letting it expand. In case of the weakly filled lattices, one may neglect interactions during the expansion, and the final spatial distribution of the atoms is given by the expectation value of the operator \(\langle \hat{\Psi}^\dagger(k) \hat{\Psi}(k) \rangle\):

\[
\langle \hat{\Psi}^\dagger(k) \hat{\Psi}(k) \rangle = |\phi(k)|^2 \sum_{m,m'} e^{i(m-m')kL} \langle a_{m'}^\dagger a_m \rangle.
\]

Thus

\[
N |\phi(k)|^2 \left( (n-|\alpha|^2) + |\alpha|^2 \sum_q \delta(k-q\frac{2\pi}{L}) \right),
\]

where \(N\) is the (large) number of lattice sites. The sum over \(q\) gives rise to a comb at lattice momenta \(q\frac{2\pi}{L}\) with a modulation proportional to the square of the mean field amplitude. This comb sits on top of a flat background caused by the incoherent population of the wells, and the whole distribution is comprised within the width of the single well momentum distribution \(|\phi(k)|^2\).

A diffraction pattern in the molecular distribution is the clear signature of a molecular condensate. By counting the released atoms or the molecules on a position sensitive detector the values of \(n\) and \(\alpha\) and thus the condensate fraction can be determined.

In summary, we have presented a method which by photoassociative removal of atoms can at the same time return an atomic system to a state with a macroscopic population of a single quantum state and prepare a molecular system with a similar macroscopic population. Weak fields owe their mean amplitude to the existence of both vacuum and single quantum excitations, and the molecular condensate exists due to the molecular vacuum component being already populated in the atomic condensate.

Eq. (8) relates the diffraction pattern to the mean field amplitudes existing prior to the detection. Two independently prepared condensates also show interference \([12]\).

It has been demonstrated theoretically, \([3]\), that this interference builds up as a consequence of the first random

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**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 is shown, starting at time zero with a coherent atomic state with unit mean number.
detection events, and that one can think of the state of the two condensates without initial mean fields as a statistical mixture of states with different relative phases [14], which all yield interference patterns but with different offsets. This same mechanism is not at work when atoms or molecules emerge from a large number of independent sites, since the uniform distribution of the phase variable at every site has vanishing probability to produce the regular progression of the phase over several sites needed for a high visibility diffraction pattern.

Our approach is quantum optical in nature: by carefully selected unitary operations we generate from a given initial state a final desired state of the system. This implies that the formal properties of the state vector of the system are made to the design, but the dynamical properties of the system may have only little in common with the properties usually ascribed to a Bose-Einstein condensate. Since the state is not the ground state or the thermodynamic equilibrium state under an applied Hamiltonian, it will not act in the same way as such a state under external perturbations, cf. for example the issue of superfluidity, discussed recently in a similar physical set-up as ours [15]. Our approach, however, suggests the possibility to create an approximation to the ground state or thermal equilibrium state of a given Hamiltonian, which can be subsequently turned on and in this way prepare states that might otherwise only be produced after prohibitively long time scales.

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