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Formation of a Bose condensate of stable molecules via a Feshbach resonance

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We propose and analyze an approach to convert an atomic condensate into a condensate of diatomic molecules in the rovibrational ground state. The process consists of a stimulated Raman transition dramatically enhanced by a time-dependent magnetic field that sweeps over a field-induced Feshbach resonance. For Na atoms the Raman transition probability is enhanced by seven orders of magnitude, leading to a conversion efficiency of up to 20%. The resulting condensate is expected to be as stable as the present atomic condensates. The approach shows promise as a more general process for Feshbach engineering molecular condensates from condensates of the constituent atoms/molecules.

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Bose-Einstein condensation (BEC) has long been known to be at the basis of macroscopic quantum phenomena such as superfluidity and superconductivity, but has only rather recently been experimentally realized in a pure and unambiguous form in cold dilute gases of alkali-metal atoms [1]. Since then the field of quantum gases has provided a rich playground with many experimental and theoretical advances [2]. The formation of a Bose-Einstein condensate of molecules is generally expected as a further breakthrough, with the development of coherent matter waves consisting of molecules (the molecule laser) as a possible application. Also, the ultimate control over all degrees of freedom of molecules realized in a condensate would open the prospect of a new type of chemical reaction (superchemistry [3]), where the macroscopic occupation of a single molecular quantum state gives rise to the coherent stimulation of a chemical reaction (bosonic stimulation).

Alkali-metal atoms are ideal for applying laser cooling, the usual first step in the cooling process needed for BEC, since their relatively simple level structure provides for the requisite closed level system for successive photon absorption and emission cycles. Unfortunately, due to their additional rovibrational level structure, molecules do not offer such possibilities. Efforts are therefore underway to cool molecules by other methods [4]. Doyle’s group at Harvard University cools and traps molecules in a magnetic trap inside a helium refrigerator. Meijer’s group at Nijmegen uses time-varying inhomogeneous electric fields to slow and trap molecules. Three groups have produced cold molecules by photoassociation of pairs of colliding atoms followed by spontaneous emission.

In this Rapid Communication, we propose and analyze an approach that directly leads to a stable condensate of molecules: the efficient and coherent conversion of an atomic condensate into a diatomic molecular condensate via a stimulated Raman transition, enhanced by a time-dependent magnetic field that sweeps over a field-induced Feshbach resonance [5]. A stimulated Raman transition induced by a pair of laser beams [6] has been used to bind freely moving pairs of condensate atoms into fragile weakly bound molecules [7], but is a very inefficient process to populate a stable low rovibrational state [8]. This is due to the poor Franck-Condon overlap of the relatively short internuclear distance range in such molecules and the more diffuse distribution of separations of a pair of trapped atoms. The transition is further obstructed by a quantum reflection region [9] in the interatomic distance range that atom pairs have to cross to reach short distances. For atoms moving at the nanokelvin kinetic energies typical for a condensate, this region is almost impenetrable. A dramatic increase of the transition probability by seven orders of magnitude can be achieved, however, by subjecting the condensate to a magnetic field tuned to a Feshbach resonance. Such a resonance occurs in the collision of two cold atoms with almost zero energy when a bound state with a Zeeman dependence different from that of a pair of free atoms crosses the threshold of the collisional energy range at a magnetic field $B_0$. From there it becomes a quasibound state [see Fig. 1(a)]. Without a change of energy, the system can then resonantly form the stable low rovibrational state, while a second laser causes further transfer to a molecular ground state.

FIG. 1. Graphical illustration of an alternative approach. (a) Schematic figures showing the crossing of a bound state with the atom + atom collision threshold. Depending on the direction of the field ramp, a real bound state below threshold or a quasibound state above threshold is formed. (b) Field ramp leading to formation of resonance state, followed by Raman photoassociation. The first laser transfers a pair of colliding atoms to an electronically excited state, while a second laser causes further transfer to a molecular ground state.
quasibound state during a collision. As illustrated in Fig.
1(b), this increases the penetration of the colliding atoms to
the short distance range, thus greatly improving the overlap
with the final molecular state.

Our approach to create a molecular condensate is closely
related to a recent experiment by Ketterle’s group [10],
showing a surprisingly strong decay of a Na condensate in a
magnetic field that varied rapidly over a Feshbach resonance.
The decay was explained via the above-mentioned different
Zeeman dependence: a significant field increase during the
brief existence of the quasibound state gives this state a sur-
plus energy, which after dissociation provides for a very ef-
fficient heating mechanism leading to the destruction of most
of the condensate [11,12]. The experiment we propose com-
bines the transient generation of a condensate of quasibound
diatoms by the field sweep method of Ref. [10] in an optical
trap with a simultaneous coherent stimulated Raman pulse,
inducing the further conversion to a permanent condensate of
stable molecules. The irreversibility introduced by the time-
dependent magnetic and Raman laser fields is a key element
of our scheme, which helps to prevent back dissociation via
the reverse bound-bound-free path.

We estimate the efficiency of this two-step approach by
considering the atom pairs in their quasibound state as a
molecular Bose-Einstein condensate, described by a coherent
field \(\phi_2(x,t)\) in addition to the field \(\phi_1(x,t)\) describing the
atomic condensate. Two further condensate components are
considered: the molecules in the intermediate electronically
excited state of the coherent Raman transition and the mol-
ecules in the final state, described by \(\phi_3(x,t)\) and \(\phi_4(x,t)\),
respectively. The evolution of the mixed condensate system
is described by a four-state model, governed by coupled field
equations [13,3,11]:

\[
\begin{align*}
\dot{\phi}_1 &= U_0 |\phi_1|^2 \phi_1 + 2 \alpha \phi_1^* \phi_2, \\
\dot{\phi}_2 &= \left( e_2 - \frac{i}{2} \gamma_0 \right) \phi_2 + \alpha \phi_1^2 + \frac{1}{2} \hbar \Omega_{L1} \phi_3, \\
\dot{\phi}_3 &= \left( e_3 - \frac{i}{2} \gamma_{sp} - \hbar \omega_{L1} \right) \phi_3 \!+\! \frac{1}{2} \hbar \Omega_{L1} \phi_2 \!+\! \frac{1}{2} \hbar \Omega_{L2} \phi_4, \\
\dot{\phi}_4 &= \left( e_4 - \hbar \omega_{L1} + \hbar \omega_{L2} \right) \phi_4 + \frac{1}{2} \hbar \Omega_{L2} \phi_3,
\end{align*}
\]

(1)

with uniform amplitudes \(\phi_j = \sqrt{n_j} \exp(i \theta_j)\) over the volume of
the condensate. Here, in the notation of Ref. [11], \(U_0 = 4 \pi \hbar^2 a_s / m\) is the off-resonant strength of the condensate
self-energy with \(a_s\) the background scattering length, the \(\alpha\)
terms describe the process that converts atoms into quasi-
bound molecules, and \(e_2 - (i/2) \gamma_0 = [B(t) - B_0] \Delta \mu
- (i/2) \gamma_0\) is the complex energy of the quasibound state
including its short-range width \(\gamma_0\) for decay inside the quan-
tum reflection region [11]. Similarly, \(e_3 - (i/2) \gamma_{sp}\) is the
complex excited-state energy with \(\gamma_{sp}\) the spontaneous
decay width and \(e_4\) the energy of the final bound molecular
state. The coupling between the \(\phi_3\) and \(\phi_4\) condensates is
induced by laser L1, and that between \(\phi_3\) and \(\phi_4\) by laser L2
(laser frequencies \(\omega_{L1}\), intensities \(I_{L1}\), and Rabi frequencies
\(\Omega_{L1}\), with \(i = 1\) and 2). We take the laser fields to propa-
gate in the same direction and to be in Raman resonance with
the final state: \(\hbar (\omega_{L1} - \omega_{L2}) = \epsilon_s\). In these circumstances
the atomic recoil is negligible [7]. Furthermore, we choose equal
linear laser polarizations perpendicular to the magnetic field.

As an example we focus on the situation of the experiment
in Ref. [10] and consider a condensate of Na atoms in the
lowest hyperfine state \(|f,m_f\rangle = |1, +1\rangle\). It is kept in an
optical trap and subjected to a magnetic field varied over one
of the Feshbach resonances at 853 G or 907 G in the direc-
tion of increasing field strength. The 853-G resonance is the
most favorable choice, since its decay rate \(\gamma_0 / \hbar\) is roughly a
factor of 100 smaller [11]. We therefore continue with this
choice. The Raman transition is taken to occur via the elec-
tronically excited bound state of \(0^\text{\footnotesize K}\) symmetry that connects
asymptotically to the \(2^3S_{1/2} + 3^3P_{1/2}\) dissociation limit. We
select the rovibrational ground state \((0,l) = (0,0)\) of the final
molecular state with \(S, M_s, L, M_L = (1, +1, 3, + 1)\), identical to that of the quasibound state [14]. Combin-
ing these two electronic ground states with a \((J, M_J) = (2,3, + 1)\) excited state leads to a favorable combination of
\(\Omega_{L1}\) and \(\Omega_{L2}\) Rabi frequencies.

We assume that the rovibrational relaxation of molecular
states due to atom-molecule and molecule-molecule colli-
sions has a relatively small effect compared to the \(\gamma_0\) decay.
We have verified that assumption for the collisions between
atoms and quasibound molecules by adding a term \(\hbar G |\phi_1|^2\)
to \(\gamma_0\) with \(G\) the rate coefficient. To very good approxima-
tion \(G\) is equal to the value \(4 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}\) for the 907-G
that the molecular states 3 and 4 are not susceptible to rovi-
brational relaxation.

The \(\alpha\) and \(\Omega\) terms describe coherent intercondensate ex-
changes of atom pairs and play a role analogous to the cou-
ping terms in the field equations for a coexisting mixed-
condensate system of atomic \(8^\text{\footnotesize Rb}\) hyperfine components
[15]. The values of the \(\alpha\) and \(\Omega\) parameters are obtained by a
full quantum scattering calculation for static \(B\) and static
laser intensities, taking into account the relevant subspaces
of unbound ground-state atoms, and bound excited and
ground states. All laser couplings between these subspaces
are taken into account to arbitrary order, as well as a spon-
taneous emission decay width for the excited state. We em-
phasize that Eq. (1), with the appropriate values of the pa-
rameters, reproduces the results of the full quantum
scattering calculation in the static case [16].

We first determine the Rabi frequency \(\Omega_{L1}\) of laser L1 by
setting the second laser intensity to zero, performing effec-
tively a one-color photoassociation (PA) experiment. In Fig.
2 we show the static photoassociation signal as a function of
the excited-state energy \(\epsilon_s\). The coupled-channels calcula-
tion is done for two cases: one for a magnetic-field value
close to resonance and the other off resonance. In agree-
ment with the Franck-Condon principle, the maxima and minima
correspond to the oscillations of the ground-state radial wave
function squared. Clearly, the Feshbach resonance enhances
the signal by seven orders of magnitude. Especially the
deeper $0^-$ states perform well since in a nonresonant case they have a poor Frank-Condon overlap. The PA signal can be described in a straightforward way by an analytical two-state model following from Feshbach’s resonance theory [5] for the quasibound state and the excited state. The inelastic transition probability is then given by

$$|S_{PA}|^2 = \frac{1}{4} \gamma_{sp} \gamma_{ab} \hbar^2 \Omega_{L1}^2 \left[ \lambda_2 \lambda_3 - \frac{1}{4} \hbar^2 \Omega_{L1}^2 \right] + \frac{1}{4} \hbar^2 \gamma_{sp} \lambda_2^2,$$

(2)

with $\lambda_2 = \varepsilon_2 - E$, $\lambda_3 = \varepsilon_3 - E - \hbar \omega_{L1}$, and $\gamma_{ab}/\hbar$ the long-range decay rate of the quasibound state to outside the quantum reflection region [11], which decreases strongly with decreasing collision energy. A comparison with the coupled-channels result for $|S_{PA}|^2$ allows us to derive $\Omega_{L1}$.

As can be seen in Fig. 2, the PA signal and thus $\Omega_{L1}$ show a strong decrease for deeper excited-state levels, reflecting the fast radial oscillations of the quasibound state for small $r$. For similar reasons the Rabi frequency $\Omega_{L2}$, coupling the excited state with the $(v,l)=(0,0)$ ground state, shows an increase for the deeper $\varepsilon_3$ levels. This leads us to select the $J=2$ state at $\varepsilon_3=-1346$ cm$^{-1}$ as a favorable compromise, with an associated outer turning point $r=13.7a_0$. As Fig. 2 shows, the one-color PA signal is in a Franck-Condon maximum at this value of $\varepsilon_3$. We find $\Omega_{L1} = 1.07 \times 10^6$ s$^{-1}\sqrt{I_{L1}}$ (W/cm$^2$). This is a very satisfactory result taking into account the loss rate $\gamma_0/\hbar = 0.69 \times 10^6$ s$^{-1}$.

In a similar way, the second Rabi frequency is determined by setting $I_{L1}$ and $\gamma_{sp}$ to zero. This isolates the bound-bound two-level system. We find $\Omega_{L2} = 1.12 \times 10^6$ s$^{-1}\sqrt{I_{L2}}$ (W/cm$^2$). Considering the spontaneous emission rate $\gamma_{sp}/\hbar = 6 \times 10^7$ s$^{-1}$, it should be possible to transfer a sizable fraction of the excited state into the ground state. Let us consider an experiment with equal Rabi frequencies $\Omega_{L1} = \Omega_{L2} = 34 \times 10^6$ s$^{-1}$ ($I_{L1} = 1000$ W/cm$^2$, $I_{L2} = 905$ W/cm$^2$), zero detunings, and a magnetic field ramp speed $\dot{B} = 0.31 \times 10^2$ G/μs, for which a 60% transfer of atoms to the quasibound state occurs in the MIT experiment [10–12]. We consider a tailored time dependence of the laser intensities with a sequence of stimulated Raman adiabatic passage (STIRAP) [17] Raman laser pulses. With a sequence of seven pulses, a 3% conversion efficiency can be obtained. This result can be improved [11] by turning to a negative magnetic-field ramp $\dot{B} = -0.31 \times 10^2$ G/μs, thus avoiding the $\gamma_0$ decay term in Eq. (1), since the $\varepsilon_3$ state is then below threshold. In Fig. 3 the solution of the coupled field equations (1) is shown for this case. We note that in this case the atom-molecule collisions remain as the only decay process for the $\phi_2$ decay. A conversion of about 20% of the
atomic condensate is achieved, including the (relatively small) reduction due to this decay.

Once a condensate of molecules is obtained, the remaining atoms can be removed by switching off the optical trap and replacing it by a magnetic trap. This will push out the weak-field seeking \((S, M_S) = (1, +1)\) state. After the removal of the atoms, the remaining decay rate due to molecule-molecule collisions can be reduced by an expansion of the trap potential.

During the 16 years of struggle to realize a BEC of atoms, the main obstacle was the instability due to inelastic processes in collisions. In the case of molecules, stability will certainly also be a vital issue. With the molecules produced in the \(v = l = 0\) state we avoid rovibrational relaxation, the main decay channel for the weakly bound molecules (lifetime of the order of tens of microseconds). To avoid also the inelastic exchange processes, we propose to fully stretch the electronic and nuclear spins by a weak-field rf or Raman transition. What remain are the dipolar two-body decay and the three-body recombination with rates comparable to those for atomic condensates. Interestingly, an additional single-molecule decay channel is the radiative decay to the lower singlet \((S=0)\) states induced by the electronic spin-orbit coupling. We estimate the decay rate to be small: of order \(0.01 \text{ s}^{-1}\) for Rb\(_2\) and even slower for Na\(_2\). The formation of Na\(_2\) molecules will be an improbable process with a small overlap of initial and final states. Details of the stability problem will be described elsewhere [18]. We expect the final BEC of molecules to be as stable as the present atomic condensates.

The Bose-Einstein condensation of diatomic alkali-metal molecules will open a subject of intense theoretical and experimental study. Compared to the existing atomic condensates with their internal spin states, molecular condensates will add the feature of new internal degrees of freedom of a spatial type: the rotational and vibrational excitations. It will be possible to study their interplay with the spin degrees of freedom. In this connection we note that the Feshbach + Raman approach can be used to create (superpositions of) higher rovibrational states with comparable or even better conversion rates. This should allow a stable multicomponent condensate system of a new type.

Another fascinating possibility would be to apply the above Feshbach + Raman technique to a degenerate gas of fermionic atoms in two different hyperfine components to allow \(s\)-wave collisions to take place. The freedom in the choice of experimental parameters within our scheme might enhance the formation of just the types of correlations that occur in a Bose condensate of Cooper pairs. Finally, Feshbach resonances in molecule-molecule scattering could be used also more generally in Feshbach catalyzing the binding of like and unlike molecules in a (multispecies) condensate to engineer condensates of heavier molecules.

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[8] Even the population of a weakly bound state is rather inefficient due to the proximity of the laser frequencies to atomic resonance and the excitation of the atomic condensate by the second laser [D. Heinzen (private communication)].
[16] With the relatively slow microsecond Feshbach field sweep, the depletion of the correlation function in the atomic condensate at short range plays a minor role [M. Holland et al., e-print cond-mat/0005062 (2000), and private communication].