Multi-mode dynamics of a coupled ultracold atomic-molecular system

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We analyze the coherent multi-mode dynamics of a system of coupled atomic and molecular Bose gases. Starting from an atomic Bose-Einstein condensate with a small thermal component, we observe a complete depletion of the atomic and molecular condensate modes on a short time scale due to significant population of excited states. Giant coherent oscillations between the two condensates for typical parameters are almost completely suppressed. Our results cast serious doubts on the common use of the 2-mode model for description of coupled ultracold atomic-molecular systems and should be considered when planning future experiments with ultracold molecules.

After experimental achievement of quantum degeneracy in atomic gases of bosons\textsuperscript{3} and fermions\textsuperscript{2}, several leading groups started the quest for creation of an analogous state in a gas of ultracold molecules\textsuperscript{4}. Such systems present a potentially much more challenging experimental task as there are several new degrees of freedom to be controlled. A major step towards this goal has been made by the Austin group of Heinzen\textsuperscript{4}, who photoassociated atoms in an atomic condensate into a selected internal molecular state producing for the first time molecules in the nK regime. Cold molecules are believed to be created as a transient state in experiments with Feshbach resonances as well\textsuperscript{5}. In this novel area there are only a few theoretical works analyzing the process of making ultracold molecules in atomic condensates and predicting several interesting phenomena in the mixed atomic-molecular systems. Javanainen \textit{et al.}\textsuperscript{6} in a series of papers\textsuperscript{6} analyzed the efficiency of photoassociation of an atomic condensate into its molecular counterpart using various theoretical schemes. Others predicted that the ground state of the hybrid system would have a soliton-like nature\textsuperscript{7,8} with liquid-like properties performing Josephson-like oscillations in response to a sudden variation of a magnetic field\textsuperscript{7}. Most frequently, coherent oscillations between the atomic and molecular condensates are envisaged\textsuperscript{9,3,7}. All the approaches employed so far have used a 2-mode approximation (only the condensates involved) to describe the dynamics of a coupled ultracold atomic-molecular system.

In this Letter we demonstrate that the 2-mode approach is inadequate as a description of current experiments on stimulated production of cold molecules in atomic Bose-Einstein condensates. For typical parameters the predicted oscillations between the two condensates are strongly damped due to significant population of excited atomic and molecular modes leading to a complete depletion of the initial condensate on a short time scale. The method we use, quantum-optical in spirit, can be regarded as a generalization of the classic Bogolubov approximation\textsuperscript{15}.

The second-quantized Hamiltonian for the hybrid atomic-molecular system confined in a box with periodic boundary conditions may be written in the following form:

\[
H = \int d^3r \left[ \frac{\Phi^\dagger \Phi}{2m} + \frac{\Psi^\dagger \Psi}{4m} - \frac{\Omega}{\sqrt{\hbar}} \int d^3r \left[ \Phi^\dagger \Phi^2 + \Psi \Phi^\dagger \Psi \right] + \frac{V}{2} \int d^3r \Phi^\dagger \Phi \Psi^\dagger \Psi \right],
\]

where \( \Phi \) and \( \Psi \) are atomic and molecular field operators, respectively, \( V = L^3 \) is a volume of the system (\( L \) being a size of the box), \( \Omega \) parameterizes a coupling between the two fields and \( g = \frac{4\pi \hbar}{\sqrt{m}} \) characterizes the atom-atom interactions in the low-energy, s-wave approximation (\( a \) being the scattering length and \( m \) – the mass of the atom; in fact, \( a \) is slightly changed in external, e.g. optical, fields – in present experiments this correction is small though\textsuperscript{15}). A similar Hamiltonian is used to describe a process of second-harmonic generation in nonlinear optics\textsuperscript{13}. The atom-molecule and molecule-molecule collisions are not included as their parameters in the low-energy regime are unknown. However, as can be seen from the construction of the method, their incorporation could be easily accomplished. Note that in this model a molecule is created when the positions of two atoms exactly coincide (for an example of a finite-range coupling approach see\textsuperscript{14}). The fields \( \Phi \) and \( \Psi \) are expanded in natural modes of the system – the plane waves:

\[
\Phi(r) = \frac{1}{\sqrt{V}} \sum_k \exp(-ik \cdot r) a_k,
\]

\[
\Psi(r) = \frac{1}{\sqrt{V}} \sum_k \exp(-ik \cdot r) b_k,
\]

where \( a_k \) and \( b_k \) are bosonic annihilation operators for atoms and molecules, respectively, and \( k = \frac{2\pi n}{L} \) with \( n_i = 0, \pm1, \pm2, \ldots (i = x, y, z) \). With this substitution the Hamiltonian assumes its final form:


\[
\frac{\hat{H}}{\hbar} = \xi \sum_k a_k^\dagger a_k + \frac{1}{2} b_k^\dagger b_k \\
+ \Omega \sum_{k,k'} b_k^\dagger a_{k'} a_k + b_{k'} a_k^\dagger a_{k'}^\dagger + \\
+ \frac{1}{2} g \sum_{k,k',k''} a_{k+k'} a_{k'} a_{k''} a_k,
\]

where \( \xi = \frac{\hbar \omega}{2m} \left( \frac{2\pi}{\lambda t} \right)^2 \).

After elimination of a fast time dependence with the substitution \( \{a, b\}_k = \exp(-i\xi n^2 t) \{\alpha, \beta\}_k \), the Heisenberg equations of motion for the operators \( \alpha_k \) and \( \beta_k \) acquire the following form:

\[
\alpha_k = -2i\Omega \sum_{k'} \exp(\frac{1}{2} i\xi |n - n'|^2 t) \beta_{k + k'} \alpha_{k'}^\dagger - \frac{1}{2} \left( i\xi |n - n'|^2 t \right) \beta_{k - k'} \alpha_{k'} \tag{4}
\]

\[
\beta_k = -i\Omega \sum_{k'} \exp(\frac{1}{2} i\xi |n - 2n'|^2 t) \alpha_{k - k'} \alpha_{k'}.
\]

The first hint about limitations of the 2-mode model comes from the following argument. As initially only the atomic condensate (the \( k = 0 \) mode) is populated, one might naively suspect that the coupling would primarily lead to an interconversion between atoms and molecules [8][9][10][11]. In such a case only the atomic and molecular condensate (\( k = 0 \)) modes would be macroscopically populated and therefore we replace the corresponding operators \( (\alpha_0 \text{ and } \beta_0) \) by c-numbers (classical complex fields) and set all the others to zero \( (\alpha_{k \neq 0} = 0 \text{ and } \beta_{k \neq 0} = 0) \). Then, in the absence of atomic collisions, time evolution of the amplitudes can be calculated analytically. Assuming \( \alpha_0 = r \exp(i\phi) \) and \( \beta_0 = \rho \exp(i\theta) \) with \( \phi = \phi_0 = \text{const} \) and \( \theta = \theta_0 = \text{const} \), the solution is:

\[
\alpha_0(t) = \frac{\sqrt{N}}{\cosh(\sqrt{2\Omega} t)} \exp(i\phi_0) ,
\]

\[
\beta_0(t) = -i \sqrt{\frac{N}{2}} \tanh(\sqrt{2\Omega} t) \exp(2i\phi_0) ,
\]

\[
\theta_0 = 2\phi_0 - \frac{\pi}{2} ,
\]

where \( N \), the total number of atoms, is a conserved quantity \( (N = |\alpha_0(t)|^2 + |\beta_0(t)|^2) \) – see Fig. 1. Note that the inclusion of interactions makes the 2-mode problem analytically insoluble (in particular the condition of constant phases \( \theta \) and \( \phi \) cannot be fulfilled any more). Numerical solutions are of oscillatory character [12][13] – different from [14]. They depict an interconversion between atomic and molecular condensate modes.

In the next step we calculate quantum corrections to such a 2-mode model treating the condensates (\( \alpha_0 \) and \( \beta_0 \)) as the only sources of particles. This amounts to leaving in Eqs. (1) the terms with at least one \( k = 0 \) (in the form of (3)) and neglecting the other ones. In the resulting equations only the zero-momentum as well as the \( k \) and \( -k \) atomic and molecular modes are present which allows to solve the coupled linear operator equations numerically. Their asymptotics \( (t \to \infty) \) may be investigated analytically though, yielding the following expectations values for the number operators:

\[
\langle \beta_k^\dagger \beta_k \rangle = \text{const} ,
\]

\[
\langle \alpha_k^\dagger \alpha_k \rangle = \frac{N \Omega^2}{2\lambda^2} \left[ \exp(2\lambda t) + \exp(-2\lambda t) - 2 \right] ,
\]

where \( \lambda = \sqrt{2\Omega^2 - \xi^2 n^4} \). \( \lambda \) is real for all modes with \( n < \sqrt{2\Omega^2 / \xi^2} \), which sets the number of modes whose population grows in time. As typically both \( \Omega \) and \( \xi \) are of the order of \( 10^{-2} \) Hz while \( N \sim 10^5 - 10^6 \) [14][15], corrections to the 2-mode model are divergent for many low-lying states. With the parameters used it is only for the 11-th and higher excited atomic modes that the quantum corrections are small and oscillatory (imaginary \( \lambda \)). Therefore, one is not allowed to exclude excited modes \( (k \neq 0) \) from a theoretical model [13].

![FIG. 1. Populations of the atomic (solid line) and molecular (dotted line) condensates calculated within a 2-mode model (see Eqs. (3)) and of the quantum corrections (dashed line) in the 1st excited atomic mode (see Eqs. (5)). The parameters are \( \Omega = 18.432 \) Hz, \( \xi = 71.373 \) Hz and \( N = 10^5 \).](image)

Fig. 1 presents an example of the run-away correction to the 2-mode model obtained numerically together with the classical source terms \( \alpha_0(t) \) and \( \beta_0(t) \) (note: in all plots the total populations are normalized by the total number of particles \( N \)). From Fig. 1 one immediately concludes that the 2-mode solution (3) is physically invalid for times larger than \( \sim 0.6 \) ms. To cure this problem, from now on, we will use a multi-mode model in the form of Eqs. (4).

Solution of the nonlinear operator equations (4) presents an extremely difficult task. A semiclassical approximation, however, is well justified for all except extremely low temperatures. Therefore, we replace all operators by c-number complex amplitudes. From the viewpoint of the Bogolubov method [12], such an approach
is legitimate as indeed many modes are macroscopically populated (i.e. their occupation is greater than quantum fluctuations). This way we are left with a set of nonlinear differential equations which must be solved numerically.

The first observation in the multi-mode model is that if one starts from a pure atomic condensate (the $k = 0$ mode), the 2-mode dynamics is recovered. However, even a very small occupation of excited atomic or molecular modes results in the dynamics beyond the 2-mode approach. Such a behavior resembles an initiation of superfluorescence where quantum, not thermal, fluctuations play a role. In a typical experiment, roughly 85% of the total number of atoms populate the atomic condensate whereas the rest of them is thermally distributed over excited atomic modes and we mimic such a situation in the initial conditions of our model \[17\]. All the molecular modes are initially unpopulated. Each atomic mode is assigned an initial, randomly chosen, phase. Any subsequent dynamics depends on the initial phases and, in a sense, a single simulation describes a single experimental realization. Values of the parameters in the model are \[\Omega =18.432 \text{ Hz}, \xi =71.373 \text{ Hz}, N = 10^5 \text{ and } g =0.018 \text{ Hz}(\text{the atomic mass and the scattering length are those of } ^{87}\text{Rb and the size of the box is equal to the Thomas-Fermi radius of a condensate of } N \text{ atoms in a trap with frequency of } \omega_0 =2\pi \times 80 \text{ Hz} - \text{see } [10,15]). \]

Sample results for the model with 2622 modes (maximum atomic and molecular excitation $n_k = -5, \ldots, 5, i = x, y, z$) are presented in Fig. 2. In fact the results stabilize for the number of modes exceeding 1000.

A striking discovery is that after a relatively short time both the atomic and molecular condensates (the $k = 0$ modes) are completely depleted and all particles occupy excited modes in roughly equal proportions. For the used (typical) parameters, only one oscillation in the condensate population survives – see Fig. 3.

This clearly indicates that if one wants to convert an atomic condensate to its molecular counterpart, it is necessary to precisely tailor the length of the coupling pulse so that the maximum of the molecular $k = 0$ mode is picked. If one investigates the condensate ($k = 0$) population and a sum of populations of all excited modes ($k \neq 0$) solely (like in Fig. 2), their dependence on the initial phases is negligible in a sufficiently big model (we checked this fact by setting different initial phases in our calculations). The effect is due to self-averaging caused by very many random phases present in the system. However, it can be observed in the time evolution of single modes. We emphasize that, contrary to the previous treatments \[9–11,15\], the effective losses from the condensates are completely due to a Hamiltonian evolution and not because of any phenomenologically introduced loss processes. In other words, due to an external weak coupling of many degrees of freedom, the system is effectively heated and its final equilibrium state certainly does not result from any $T = 0$ dynamics. The inclusion of atom-molecule and molecule-molecule collisions would randomize the still coherent dynamics leading to an analogous and even more pronounced effect. Remarkably, the character of the dynamics does not depend on the total number of particles – our simulations for $N = 2 \cdot 10^3$ and $N = 5 \cdot 10^5$ still show only one oscillation in the atomic condensate population on a slightly altered (longer for smaller $N$) time scale. The amplitude of this oscillation is bigger for smaller $N$ indicating a small increase in the molecular condensate production ($\sim 30\%$ for $N = 2 \cdot 10^3$ and $\sim 23\%$ for $N = 5 \cdot 10^5$ instead of $\sim 25\%$ for $N = 10^5$ as seen in Fig. 3).

In order to recover the standard oscillatory dynamics of the 2-mode limit \[9–11\] within a multi-mode model, one needs to detune the excited modes. The latter can be achieved by decreasing the size of the box (and so enlarging a spacing between the excited modes) while

\[]\[\text{FIG. 2. Populations of atomic (thick solid line) and molecular (thick dashed line) condensates (the } k = 0 \text{ modes) and the sums of populations of all excited } k \neq 0 \text{ atomic (thin solid line) and molecular (thin dashed line) modes in the model with 2622 states.}\]\[\text{FIG. 3. 2-mode (dashed line) vs. multi-mode (solid line) dynamics of an atomic condensate population.}\]
keeping the total density fixed (in order not to alter the scattering and coupling parameters). The results for the box whose volume is $15^3$ times smaller are presented in Fig. [4] (the appropriately decreased number of particles is $N = 10^9/15^3 \simeq 30$).

To summarize, we point out the invalidity of a 2-mode model for a description of present experiments with coupled atomic-molecular systems in the Bose-Einstein condensation regime. We find it necessary to employ a multimode approach and within it we observe a complete depletion of both the atomic and molecular condensates on a short time scale. For typical parameters, only one giant oscillation between the two condensates is present. Thus, a new destructive mechanism in the system is pointed out which should be taken into account while planning future experiments. An effectively 2-mode dynamics is recovered under special conditions of large detunings. The system presents an interesting example of a quantum coherent dynamical evolution effectively randomized by the coupling of many degrees of freedom.

In this Letter we presented the calculations for the box rather than the harmonic oscillator potential. We have chosen the box size such that the first excitation energy is of the order of the level spacing in the experiment of Heinzen [4]. Hence, we may expect that for the harmonic oscillator the time of coherent evolution would be even shorter, because of the quadratic vs. linear dependence of excitation energies in the respective potentials.

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16. At the time of writing this manuscript we found out that the problem of going beyond the 2-mode dynamics had been also addressed in M. Holland, J. Park, and R. Walser, cond-mat/0005062. The authors point out an effect of condensate depletion due to a thermal cloud. However, in their treatment they neglect all collisions.
17. We do recover the 2-mode dynamics in the semiclassical approximation starting from the pure atomic condensate (very low temperature). However, as we see in Fig. [4] even in the case of a 100% initial occupation of the atomic condensate, the quantum initiation makes the 2-mode model invalid after $\sim 0.6$ ms.