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Coherent photoassociation of a Bose-Einstein condensate

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We propose a phenomenological Hamiltonian for the analysis of photoassociation of a Bose-Einstein condensate. We find that, by adiabatically sweeping the frequency of the driving laser, a condensate of atoms may be converted rapidly to a condensate of molecules. [S1050-2947(99)50905-0]

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In photoassociation (PA), (two) atoms combine in the presence of light to make a (diatomic) molecule. PA of cold atomic gases has served the currently prominent studies of Bose-Einstein condensation (BEC) in alkali-metal vapors [1] mainly by providing scattering length data [2]. Recently, a more intimate entanglement of BEC and PA has been brought to the fore by the suggestions that it may be possible to produce a Bose-Einstein condensate of molecules by photoassociating a condensate of atoms [3,4]. We have, in fact, argued that the density parameter that determines the steady-state PA yield is essentially the same phase-space density that governs condensation [4]. However, past theoretical analyses of PA of cold atoms [3–7] do not explicitly take into account the statistics of the atoms and the molecules. Besides, studies of a thermal gas invariably boil down to adding up PA rates, as opposed to amplitudes, due to the individual colliders. Consequently, coherent processes such as stimulated Raman adiabatic passage (STIRAP) [7] are predicted to be unimportant [4].

In this Rapid Communication we propose a model Hamiltonian that accounts for the statistics of both the atoms and the molecules. In the limit of nondegenerate gas we recover the standard approach [3–7]. In a degenerate gas, though, coherent processes may be prominent. As examples we discuss both analytically and numerically nonlinear oscillations in which atoms turn into molecules and back, and an adiabatic process in which the majority of the atoms is transformed into molecules with a sweep of the laser frequency.

According to the prevailing picture, PA is seen as a free-bound transition from the dissociation continuum of a diatomic molecule to a bound state. Nevertheless, in a container of gas there are typically many more than two atoms. In a description of PA of a condensate two issues arise, both in a sense because one cannot unambiguously group atom pairs into molecules. First, as we have mentioned already, one calculates the PA rate for a pair of atoms and then multiplies (implicitly or explicitly) the result by the number of colliders to obtain the total PA rate. We wish to avoid such a rate approximation. Second, both the atoms and the molecules should be described as bosons. The statistics of atoms and molecules in terms of the statistics of their constituents is the usual question; that the atoms and molecules commingle and may be converted into one another adds the extra twist.

We bypass these issues by adopting a phenomenological Hamiltonian. For notational simplicity we take all atoms to be in the same internal state, and the same with the molecules. We thus have to consider c.m. motion only. For the time being we assume that both the atoms and the molecules make free ideal gases quantized in a cubic box of volume V, and use the wave vectors $\mathbf{k}$ to characterize the motion. We denote by $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ the boson annihilation operators for atoms and molecules. The corresponding eigenfrequencies are $\epsilon_{\mathbf{k}}=\hbar\mathbf{k}^2/2m$ and $\varepsilon_{\mathbf{k}}=\hbar\mathbf{k}^2/4m$ for atoms and molecules with the respective masses m and $2m$. We assume that a plane wave of light with the electric field amplitude $\mathbf{E}_0$ and wave vector $\mathbf{q}$ drives a dipole transition that seeks to glue two atoms into a molecule by effecting a transition from the dissociation quasicontinuum [4] to a higher-lying vibrational bound state of the molecule. We denote by δ the detuning of the laser, such that $\hbar\delta$ gives the energy of the state above the dissociation threshold that is on light-driven resonance with the bound molecular state. The Hamiltonian reads

$$\frac{H}{\hbar}=\sum_{\mathbf{k}}\left[\varepsilon_{\mathbf{k}}b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}}+\left(-\frac{\delta}{2}+\epsilon_{\mathbf{k}}\right)a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\right]-\sum_{\mathbf{k}\mathbf{k}'}\left[\frac{\mathbf{d}_{\mathbf{k}\mathbf{k}'}\cdot\mathbf{E}_0}{4\hbar}b_{\mathbf{k}+\mathbf{k}'+\mathbf{q}}^{\dagger}a_{\mathbf{k}}a_{\mathbf{k}'}+\text{H.c.}\right]. \tag{1}$$

Momentum conservation is explicitly built into Eq. (1), in that two atoms with momenta $\hbar\mathbf{k}$ and $\hbar\mathbf{k}'$ plus a photon with momentum $\hbar\mathbf{q}$ combine to make a molecule with momentum $\hbar(\mathbf{k}+\mathbf{k}'+\mathbf{q})$. The precise nature of the dipole moment matrix elements $\mathbf{d}_{\mathbf{k}\mathbf{k}'}$ is yet to be determined. For the time being we simply assume that $\mathbf{d}_{\mathbf{k}\mathbf{k}'}=\mathbf{d}_{\mathbf{k}'\mathbf{k}}$.

To make the connection with the conventional models of PA [3–7], we momentarily drop photon recoil from the Hamiltonian by setting $\mathbf{q}=0$. Because momentum is conserved, only atom pairs such that $\mathbf{k}+\mathbf{k}'=\mathbf{K}$ couple to a molecule with the wave vector $\mathbf{K}$, and every total momentum $\hbar\mathbf{K}$ may be treated separately. In a given $\mathbf{K}$ manifold we define the relative momentum of the colliding atoms $\hbar\mathbf{p}$ in such a way that $\mathbf{k}=\mathbf{K}/2+\mathbf{p}$, $\mathbf{k}'=\mathbf{K}/2-\mathbf{p}$. We assume that only two atoms are present with this total momentum $\hbar\mathbf{K}$. The dipole coupling therefore connects the state $|\mathbf{K}\rangle$, with the molecule in the state $\mathbf{K}$ and no atoms, only to the manifold of states $|\mathbf{p}\rangle$ with no molecule and one atom in each of the states $\mathbf{K}/2\pm\mathbf{p}$. We may write the state vector as $|\psi\rangle=\Sigma_{\mathbf{p}}\alpha_{\mathbf{p}}|\mathbf{p}\rangle+\beta|\mathbf{K}\rangle$, and express the time-dependent Schrödinger equation in terms of the expansion coefficients $\alpha_{\mathbf{p}}$ and β as

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$$\dot{\alpha}_{\mathbf{p}} = -i\left[\varepsilon_{\mathbf{K}} + \frac{\hbar \mathbf{p}^2}{m} - \delta\right]\alpha_{\mathbf{p}} + \frac{i\mathbf{E}_0^*}{2\hbar}\cdot \mathbf{d}^*_{\mathbf{K}/2-\mathbf{p},\mathbf{K}/2+\mathbf{p}}\beta, \tag{2a}$$

$$\dot{\beta} = -i\varepsilon_{\mathbf{K}}\beta + \frac{i\mathbf{E}_0}{2\hbar}\cdot\sum_{\mathbf{p}} \mathbf{d}_{\mathbf{K}/2-\mathbf{p},\mathbf{K}/2+\mathbf{p}}\alpha_{\mathbf{p}}. \tag{2b}$$

For two bosons the states $|\mathbf{p}\rangle$ and $|-\mathbf{p}\rangle$ are actually the same. Accordingly, we may only use half of the possible **p** values in Eqs. (2), say, those with $p_x > 0$.

We now assert the equality $\mathbf{d}_{\mathbf{k}\mathbf{k}'} = (1/\sqrt{2})[\mathbf{d}(\mathbf{p}) + \mathbf{d}(-\mathbf{p})]$, where $\mathbf{d}(\mathbf{p})$ is the dipole moment matrix element between the bound state of the molecule and the dissociated state that asymptotically (at large distances) corresponds to the two atoms having the relative momentum **p**. This completes our model.

A full analysis of statistics and degeneracy is presumably only necessary at very low temperatures. It is therefore reasonable to assume that only *s*-wave collisions have to be considered, which we do throughout the rest of this paper. Then $\mathbf{d}(\mathbf{p})$ is a function of $|\mathbf{p}|$ only, and $\mathbf{d}_{\mathbf{k}\mathbf{k}'} = \sqrt{2}\,\mathbf{d}(\mathbf{p})$. By comparing with the quasicontinuum approach to PA [4], it may be seen that Eqs. (2) are equivalent to the standard analysis of PA (and photodissociation) for two atoms, except that in Eqs. (2) the matrix element responsible for PA is larger by a factor of $\sqrt{2}$ and that there is the restriction on the relative momenta **p**.

The differences are explained when we consider a nondegenerate thermal gas. States with more than two atoms for any given relative motion **p** are then rare, so that Eqs. (2) should be an essentially complete description of the physics for each state **K** of the photoassociated molecule. Moreover, different states of relative motion are initially uncorrelated, so that one adds probabilities, not amplitudes, for PA transitions starting from each **p**. Probability is proportional to the square of the matrix element, so for each **p**, Eqs. (2) give a rate that is twice the rate of the standard approach. In balance, in the conventional treatment there is no restriction of **p** vs $-\mathbf{p}$, so one adds over twice as many **p** as here. In the end, Eqs. (2) and the usual analysis give the same results.

Things change when one starts out with a condensate with all N atoms in the state $\mathbf{k}=0$. The primary light-induced coupling is to the molecular state $\mathbf{K}=\mathbf{q}$, and for the time being we only take into account the two modes $a \equiv a_0$, $b \equiv b_{\mathbf{q}}$. Furthermore, we drop a term proportional to the conserved quantity $a^\dagger a + 2\,b^\dagger b$, and obtain the two-mode Hamiltonian

$$\frac{H}{\hbar} = -\frac{\delta'}{2}a^\dagger a - \frac{\kappa}{2}(b^\dagger a a + b a^\dagger a^\dagger). \tag{3}$$

Here $\delta' = \delta + \varepsilon_{\mathbf{q}}$ is the detuning corrected for photon recoil of the molecule, and κ is the Rabi frequency, $\sqrt{2}$ times the one we introduced in Ref. [4].

The Hamiltonian (3) is the model that has been employed for second-harmonic generation since the early days of quantum optics. At this point the relevant literature is extensive; for a few snapshots see Refs. [8–10]. Our first example on coherent PA is a phenomenon predicted long ago [8], though to our knowledge never seen experimentally. Namely, suppose that the effective detuning is set equal to zero, $\delta' = 0$. In the conventional two-level atom on exact resonance one finds Rabi flopping: the atom is transferred between the two states at the frequency 2κ. Here similar flopping should ensue. To find the time scale, we write the Heisenberg equation of motion for, say, the operator a, which gives $\dot{a} = i\kappa\, a^\dagger b$. If the system starts out with N atoms, the operators a and b presumably have the characteristic size $\sqrt{N}$. In terms of operators that are of the order of unity, $A = a/\sqrt{N}$ and $B = b/\sqrt{N}$, the equation of motion becomes $\dot{A} = i(\sqrt{N}\kappa)A^\dagger B$. The principal frequency scale for the evolution should be $\Omega = \sqrt{N}\kappa$.

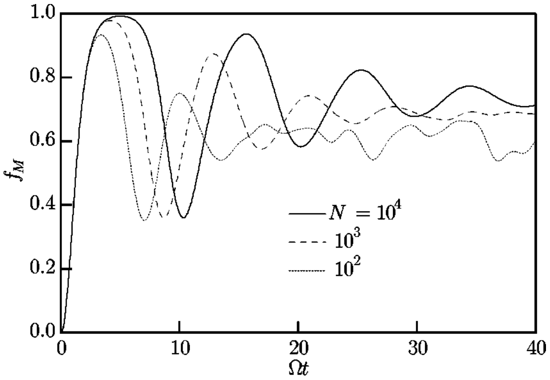

FIG. 1. Fraction of atoms converted to molecules $f_M = 2\langle b^\dagger b\rangle/N$, as a function of dimensionless time Ωt, for on-resonance oscillations between a condensate of atoms and a condensate of molecules. The curves for atom numbers $N=10^2$, 10^3, and 10^4 are identified in the legend.

This indeed is the case, as we demonstrate by integrating the time-dependent Schrödinger equation numerically. In Fig. 1 we plot the fraction of atoms converted to molecules f_M as a function of the dimensionless time parameter $\tau = \Omega t$, starting with $N=10^2$, 10^3, and 10^4 atoms. Nonlinear oscillations on a time scale $\Delta\tau \sim 1$, or $\Delta t \sim 1/\Omega$, are seen. The oscillations are damped away, the more slowly (in τ), the larger the atom number. At longer times still, the oscillations periodically revive [10].

One should evidently clarify what the frequency scale $\Omega = \sqrt{N}\kappa$ means in experimental terms. It was argued in [4] that the Rabi frequency κ tends to zero with growing quantization volume V as $V^{-1/2}$. Nonetheless, if at the same time the number of condensate atoms N tends to infinity in such a way that the condensate density $\rho = N/V$ remains constant, the frequency Ω has a finite continuum limit for $V \to \infty$. Using the same state counting procedure as before [4], we first express Ω in terms of the density ρ, the rate of photodissociation (PD) of the ensuing molecules in the given laser light Γ, the relative velocity of colliding atoms v, and the reduced mass $\mu = m/2$, as

$$\Omega = \sqrt{N}\kappa = \sqrt{2\pi\hbar^2\Gamma\rho/\mu^2 v}. \tag{4}$$

In our BEC model the collision velocity is $v=0$, but for *s*-wave collisions the PD rate Γ also tends to zero at the threshold as $\Gamma \propto v$, and the expression Γ/v behaves well in the limit $v \to 0$.

Using the relation (4), any calculation or measurement of the PA or PD rate at a low enough temperature may be turned into a prediction of the frequency Ω. Here we adopt the analysis of Ref. [6] to develop an *ab initio* estimate. In the limit of separated atoms, there is an underlying atomic transition for the PA transition. It turns out that the characteristic frequency Ω may be related to the (on resonance) Rabi frequency Ω_A that the same laser would drive in the atomic transition. Up to various numerical constants that depend on the details of the molecular structure, generally of the order of unity [11], the result is

$$\Omega \sim \sqrt{\lambda\!\!\!^{-3}\rho}\,\sqrt{\nu\gamma_A/\Delta^2}\,\Omega_{\rm A}. \tag{5}$$

Here $\lambda\!\!\!^{-}$ is given terms of the wavelength of the driving light λ as $\lambda\!\!\!^{-}=\lambda/(2\pi)$, ν and $\hbar\Delta$ are the vibration frequency and the binding energy of the photoassociated state of the molecule, and γ_A is the spontaneous decay rate for the underlying atomic transition.

For a Bose-Einstein condensate it may well happen that $\sqrt{\lambda\!\!\!^{-3}\rho}\sim 1$, and for a highly excited molecular state the other square-root factor in Eq. (5) may also be of the order of unity. Characteristic frequencies Ω reaching all the way up to the Rabi frequency Ω_A driven by the same laser in a parent atom are not out of the question.

In order to prepare for our final topic, we momentarily return to the Hamiltonian (1). We represent atoms and molecules using the respective boson fields $\phi(\mathbf{r}) = V^{-1/2}\Sigma_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}}$, $\psi(\mathbf{r}) = V^{-1/2}\Sigma_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} b_{\mathbf{k}}$, and express the dipole matrix elements $\mathbf{d}_{\mathbf{kk}'}$ in terms of a function $\mathbf{d}(\mathbf{r})$ defined by

$$\mathbf{d}_{\mathbf{kk}'} = \frac{1}{\sqrt{V}}\int d^3r\, e^{[i(\mathbf{k}-\mathbf{k})/2]\cdot\mathbf{r}}\,\mathbf{d}(\mathbf{r}). \tag{6}$$

By considering the scaling of the matrix elements with the quantization volume V once more [4], it may be seen that $\mathbf{d}(\mathbf{r})$ does *not* depend on V. The Hamiltonian (1) is the integral over the position $\mathbf{r}$ of the Hamiltonian density

$$\begin{aligned}\frac{\mathcal{H}(\mathbf{r})}{\hbar} &= \psi^\dagger\left(-\frac{\hbar\nabla^2}{4m}\right)\psi + \phi^\dagger\left(-\frac{\delta}{2}-\frac{\hbar\nabla^2}{2m}\right)\phi \\ &\quad -\frac{1}{2\hbar}\bigg[\mathbf{E}^+\cdot\psi^\dagger\int d^3r'\,\phi(\mathbf{r}+\tfrac{1}{2}\mathbf{r}') \\ &\quad \times\mathbf{d}(\mathbf{r}')\,\phi(\mathbf{r}-\tfrac{1}{2}\mathbf{r}')+\text{H.c.}\bigg].\end{aligned} \tag{7}$$

We have omitted the position argument of a field if it is the default $\mathbf{r}$. $\mathbf{E}^+(\mathbf{r}) = \frac{1}{2}\mathbf{E}_0 e^{i\mathbf{q}\cdot\mathbf{r}}$ stands for the positive frequency part of the driving laser light.

The virtue of the expression (7) is that it is independent of the bases used to describe atomic, molecular, and optical fields. As called for by the physics of the problem on hand, one may add confining potentials for atoms and molecules, add collisional interactions between various particles, accommodate for the non-plane-wave shape of the electric field, develop approximations akin to the Gross-Pitaevskii equation, and so forth. This said, an experimental problem with the oscillations in Fig. 1 is obvious. Due to atom-atom and molecule-molecule interactions, the energy of a state with given numbers of atoms and molecules depends nonlinearly on the numbers. In practice one cannot maintain exact resonance by simply keeping the laser frequency fixed.

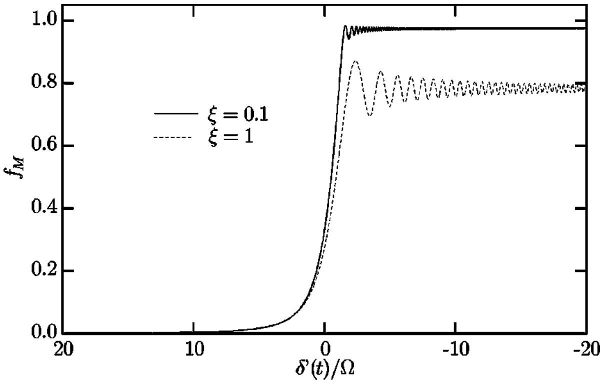

FIG. 2. Fraction of atoms converted to molecules f_M, as a function of instantaneous detuning $\delta'(t)$, when the detuning is swept linearly as a function of time according to $\delta'(t) = -\xi\Omega^2 t$. The figure is for the atom number $N=100$, for the two sweep rates $\xi = 0.1$ and $\xi = 1$.

Adiabaticity offers excitation methods that are more robust than straight resonant drive. As our second example of coherent PA we discuss a version of adiabatic following [12], see also [13], using the two-mode model (3). Starting with all atoms and a large positive detuning, one sweeps the detuning across the resonance [14]. For large positive detunings the ground state of the Hamiltonian (3) is the state with all atoms, for large negative detunings it is the state with all molecules. If the detuning is swept ''slowly enough,'' the system will move adiabatically from the state with all atoms to the state with all molecules.

We study the linear frequency sweep $\delta'(t) = -\xi\Omega^2 t$, where ξ is a dimensionless parameter characterizing the rapidity of the sweep. If Ω is the applicable frequency scale, adiabaticity should hold once $\xi \ll 1$. A corroboration is shown in Fig. 2. We again solve the Schrödinger equation numerically, and plot the fraction of atoms converted to molecules as a function of the instantaneous detuning $\delta'(t)$. The sweep rates are $\xi=0.1$ and $\xi=1$, and the initial atom number is $N=100$. The system is taken as starting in the eigenstate of the Hamiltonian (3) pertaining to the initial detuning $\delta' = 20\Omega$. The molecular yield greatly improves and the relaxation oscillations become much smaller upon reducing ξ from 1 two 0.1. Direct dependence of adiabaticity on the atom number N, other than through the parameter Ω, was found to be weak for $N \gg 1$.

Some words of caution regarding our examples are due. We have so far said little about PD. In the picture of our earlier paper [4], PD entails effectively irreversible transitions from the bound molecular state to the quasicontinuum of the dissociated molecule. If the final states of PD (as opposed to the one state containing the condensate) have small occupation numbers, PD proceeds similarly and at the same rate (per molecule) as it would in a nondegenerate gas.

In our example about atom-molecule oscillations, the light is tuned to the threshold of PA *and* PD. This implies that the PD rate is nominally $\Gamma = 0$, and PD should be unimportant. In the case of adiabatic following, the frequency sweep starts from above the dissociation threshold, but initially there are no molecules to photodissociate. As may be seen from Fig.

2, a significant population of molecules emerges only when $\delta' \simeq \Omega$. Let us denote the PD rate for this laser tuning by Γ_0, and the corresponding relative velocity of the molecular fragments by v_0; evidently $\mu v_0^2/2 = \hbar\delta' \simeq \hbar\Omega$. After another time interval $\simeq (\xi\Omega)^{-1}$, $\delta' = 0$ and the PD channel closes for good. Using Eq. (4), the fraction of atoms lost to PD during the adiabatic sweep may thus be estimated as

$$\frac{\Gamma_0}{\xi\Omega} \simeq \frac{1}{8\sqrt{2}\,\pi}\,\frac{1}{\xi}\,\frac{1}{\lambdabar^3\rho}\left(\frac{\Omega}{\epsilon_R}\right)^{3/2}. \tag{8}$$

Here $\epsilon_R = \epsilon_{\mathbf{q}} = \hbar/(2m\lambdabar^2)$ is the conventional recoil frequency for laser cooling, of the order of $2\pi \times 100$ kHz for the *D* lines of alkali metals [14]. While one might be tempted to expedite adiabatic transfer with increased laser intensity, Eq. (8) tells us that increased PD losses are eventually the price. Taking qualitatively $\xi \sim 1$, $\lambdabar^3\rho \sim 1$, the fastest one can carry out an efficient atom-molecule conversion is in a time $\sim \epsilon_R^{-1} \sim 1$ μs.

We have assumed free atoms and molecules, while in the present-day experiments a likely state of affairs would be trapped atoms and free molecules [1]. Under the prevailing experimental conditions the characteristic time scales of both trapped atoms (inverses of trap frequencies) and free molecules (ballistic expansion time, the time it takes molecules to fall from the trap under gravity) are at least of the order of 1 ms. For time scales shorter than this, such as the conceivable 1-μs time scale for atom-molecule conversion, trapping or the absence thereof should not make much difference. Pending a detailed investigation of trapping using a field theory along the lines of Eq. (7), there may be an experimental window even for our simplest free-atom, free-molecule arguments.

There are other experimental complications we have not addressed. For instance, a molecule that is created in an electronically excited state by means of a dipole transition also decays spontaneously at a rate comparable to the atomic emission rate γ_A, and the result need not be two atoms back to the condensate. Three-level schemes [7,3] have been devised to counteract such spontaneous-emission loss. Our present approach, too, could easily accommodate as many internal states of the molecule as desired. Besides, with a condensate initial state, our objection [4] against STIRAP [7] in PA is moot. It could be that STIRAP works in PA starting with a condensate.

In sum, we have proposed a model Hamiltonian for the analysis of photoassociation of a Bose-Einstein condensate. We have, in a way, added second-harmonic generation to the arsenal of ''nonlinear atom optics'' [15]. As introductory examples we have demonstrated coherent transitions, atom-molecule oscillations and adiabatic following, between atomic and molecular condensates, and analyzed the relevant frequency/time scale as a function of the intensity of the driving laser. We have also cast our approach in terms of a general field theory. While much work is needed to clarify the effects of practical experimental circumstances on coherent photoassociation, our model should readily lend itself to such studies.

Note added in proof. Recently closely related work by Drummond *et al.* has appeared. See, in particular, Ref. [16].

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