Dissociation dynamics of a Bose-Einstein condensate of molecules

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An unstable condensate of diatomic molecules will coherently disassociate into correlated pairs of atoms. This dissociation process exhibits very rich quantum dynamics depending on the quantum statistics of the constituent atoms. We show that in the case of bosonic atoms Bose enhancement can lead to stimulated dissociation, whereas, in the case of fermions Pauli blocking of the available states and a buildup of coherence between molecules and atom pairs can give rise to incomplete dissociation of the molecules and transient association-dissociation oscillations.

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The ability to create quantum degenerate molecules composed of fermionic [1–3] or bosonic [4,5] atoms by tuning a molecular level into resonance with the atomic states via a Feshbach resonance [6–8] or by photoassociation [9,10] has opened up an exciting new area of physics for exploration. The case of diatomic bosonic molecules coupled to bosonic atoms ($b \leftrightarrow bb$) has been shown to undergo coherent association-dissociation oscillations [11] and there are predictions of Bose-enhanced phenomena in this system that may lead to a, so called, superchemistry [12–15]. The case of diatomic bosonic molecules coupled to fermionic atoms ($b \leftrightarrow ff$) has also received a lot of attention lately due to the possibility of realizing a BEC-BCS crossover [16–18]. For positive detuning from resonance (where two-body theory predicts unstable molecules) molecules can be stabilized by Pauli-blocking of the atomic states [19] and coherent population oscillations have also been predicted to occur in this case [20–22]. In these systems quantum statistics obviously play an important role and it is becoming clear that atom-molecule coherence generated by their coupling is one of the key elements to understanding their equilibrium and nonequilibrium behavior [20,22–24].

In this paper we consider the production of correlated pairs of atoms by the spontaneous dissociation of a pure Bose-Einstein condensate (BEC) of molecules [25–28]. This highly nonequilibrium, spontaneous-dissociation regime can be reached by first creating a stable BEC of molecules far from the resonance, then rapidly tuning through to the other side of the resonance, i.e., $\nu<0 \rightarrow \nu>0$, where ν is the detuning of the molecular level from the atomic continuum and depends on the applied magnetic field in the case of a Feshbach resonance or the detuning of the coupling laser field in the case of photoassociation-dissociation. Once the molecule level is above the atomic continuum the molecules will become unstable and begin to dissociate into atomic pairs. For a condensate in a zero momentum state the atoms are created in correlated pairs of equal and opposite momentum centered at $k=\sqrt{2m\nu/\hbar}$. Correlated pair production by this method has been discussed previously in the case of bosonic atoms [12,13] and recently also in the case of fermionic atoms [29]. Here we present a unified treatment of the dissociation dynamics of a molecular condensate which includes both the boson ($b \rightarrow bb$) and fermion ($b \rightarrow ff$) case.

Assuming the momentum spread of the molecular condensate is negligible compared to the mean momentum of the emitted atoms, the Hamiltonian of the system can be approximated by

$$H = \hbar \nu a_0^\dagger a_0 + \hbar \sum_{\boldsymbol{k}} \omega_{\boldsymbol{k}} (b_{\boldsymbol{k}\uparrow}^\dagger b_{\boldsymbol{k}\uparrow} + b_{\boldsymbol{k}\downarrow}^\dagger b_{\boldsymbol{k}\downarrow}) + \hbar g \sum_{\boldsymbol{k}} (a_0^\dagger b_{-\boldsymbol{k}\downarrow} b_{\boldsymbol{k}\uparrow} + a_0 b_{\boldsymbol{k}\uparrow}^\dagger b_{-\boldsymbol{k}\downarrow}^\dagger), \quad (1)$$

where a_0 is the bosonic molecular mode and $b_{\boldsymbol{k}\uparrow}$ and $b_{\boldsymbol{k}\downarrow}$ are the annihilation operators of the atoms and either satisfy bosonic or fermionic commutation relations. $\omega_{\boldsymbol{k}}=\hbar k^2/2m$ is the dispersion relation of the atoms and g is the coupling between the closed channel (molecules) and the open channel (free atoms) of the coupled channels scattering problem [6]. We have assumed that the atoms are in different internal states denoted by $\uparrow$ and $\downarrow$ but the conclusions can be straightforwardly applied to the case of only one bosonic atomic species. The total number of atoms, $N=2a_0^\dagger a_0+\Sigma_{\boldsymbol{k}}(n_{\boldsymbol{k}\uparrow}+n_{\boldsymbol{k}\downarrow})$ is conserved, where $n_{\boldsymbol{k}s}=b_{\boldsymbol{k}s}^\dagger b_{\boldsymbol{k}s}$ is the number operator for the atoms. As atoms of opposite spin and momentum are created and destroyed as pairs, the number difference, $n_{\boldsymbol{k}\uparrow}-n_{-\boldsymbol{k}\downarrow}$, is also conserved. In the boson case the correlation between the atoms created via molecular dissociation is analogous to that between photons created in a non-degenerate parametric amplifier (see [30] and references within). Quite recently, the pair correlations between fermionic atoms with opposite momenta have been observed in the noise spectrum of photodissociated cold molecules [31], following the theoretical proposal of Ref. [32]. This pair correlation does not require the presence of a molecular condensate as it is simply a consequence of the form of the Hamiltonian and arises even in an incoherent dissociation process (such as from thermal molecules). On the other hand, only coherent molecular dissociation, such as from a condensate, can give rise to atom-molecule coherence and the related phenomena of coherent association-dissociation oscilations [11]. This coherence—characterized by a nonzero value of $\langle a_0^\dagger b_{-\boldsymbol{k}\downarrow} b_{\boldsymbol{k}\uparrow}\rangle$—plays an important role in the present work.

Given that the atoms are created in pairs, we can take advantage of a formal mapping between pairs of fermion operators and spin-1/2 Pauli matrices to write the Hamil-

tonian in a more natural form. This mapping has been exploited to determine the phase diagram of the BCS-BEC crossover and to predict non-equilibrium atom-molecule oscillations [20–22]. A similar mapping can be made for boson pairs and we treat the two cases in parallel. We define new operators by $\sigma_{k-}=b_{-k\downarrow}b_{k\uparrow}$, $\sigma_{k+}=b^{\dagger}_{k\uparrow}b^{\dagger}_{-k\downarrow}$ and $\sigma_{kz}=\frac{1}{2}(n_{k\uparrow}+n_{-k\downarrow}\mp 1)$. It is easy to check that these operators satisfy the commutation relations: $[\sigma_{k\pm},\sigma_{k'\pm}]=0$, $[\sigma_{kz},\sigma_{k'+}]=\delta_{k,k'}\sigma_{k+}$, $[\sigma_{kz},\sigma_{k'-}]=-\delta_{k,k'}\sigma_{k-}$ and

$$[\sigma_{k+},\sigma_{k'-}]=\pm 2\delta_{k,k'}\sigma_{kz}, \tag{2}$$

where the upper (lower) sign in Eq. (2) corresponds to fermions (bosons). The seemingly insignificant sign difference in the commutation relations in the two cases leads to completely different dynamics.

Writing the Hamiltonian (1) in terms of these new operators we have (minus a constant)

$$H=\hbar\sum_{k}[2(\omega_{k}-\nu)\sigma_{kz}+g(a_{0}^{\dagger}\sigma_{k-}+\sigma_{k+}a_{0})], \tag{3}$$

and $N=2a_{0}^{\dagger}a_{0}+\Sigma_{k}(2\sigma_{kz}\pm 1)$. In the fermion case, this Hamiltonian describes an ensemble of independent two-level systems interacting with a single bosonic mode. The case of identical two-level systems: $\omega_{k}=\omega_{0}$, is called the Dicke model and is an exactly solvable model that has been extensively studied in the quantum optics literature (see Ref. [33] and references within).

In the special case where the population in each mode is small, ($\langle n_{k\uparrow}+n_{k\downarrow}\rangle\ll 1$), throughout the dissociation process, due to a large number of available states, we can make the approximation that $[\sigma_{k-},\sigma_{k'+}]\approx\delta_{k,k'}$, independent of whether the operators describe bosons or fermions. In other words, the underlying quantum statistics of the atoms are unimportant and the Hamiltonian describes the coupling of a single mode to a continuum of bosonic modes with a quadratic dispersion relation. This model has been studied previously in the context of an atom laser [34]. Under the Born-Markov approximation, which holds for weak coupling and large ν [35], the molecules will experience a rather trivial exponential decay and the final frequency distribution of the atom pairs will be the standard Lorentzian.

To proceed further in the general case we note that we are interested in the spontaneous dissociation of an initially large molecular condensate, so we make a mean-field approximation for the molecular mode by replacing the operator $a_{0}(t)$ with a c-number $\alpha(t)$ (taken to be real without loss of generality). This approximation will break down when the population of molecules approaches zero and quantum fluctuations of the molecular mode become important. Under the mean-field approximation, the atom-molecule coherence reduces to $\langle a_{0}^{\dagger}b_{-k\downarrow}b_{k\uparrow}\rangle=\alpha\langle b_{-k\downarrow}b_{k\uparrow}\rangle=\alpha\langle\sigma_{k-}\rangle$ and the equations of motion for the averages $\langle\sigma_{ks}\rangle$ can be written as

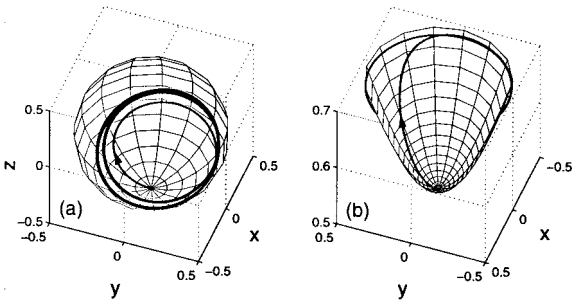

FIG. 1. Example trajectories of $\boldsymbol{S}_{k}(t)$ for fermions (a) and bosons (b).

$$\frac{d\boldsymbol{S}_{k}}{dt}=\begin{pmatrix}0 & -2(\omega_{k}-\nu) & 0\\ 2(\omega_{k}-\nu) & 0 & \mp 2g\alpha(t)\\ 0 & 2g\alpha(t) & 0\end{pmatrix}\boldsymbol{S}_{k}, \tag{4}$$

where $\boldsymbol{S}_{k}=[\langle\sigma_{kx}\rangle,\langle\sigma_{ky}\rangle,\langle\sigma_{kz}\rangle]^{t}$ is the column vector of averages and we have defined $\sigma_{kx}=(\sigma_{k+}+\sigma_{k-})/2$ and $\sigma_{ky}=(\sigma_{k+}-\sigma_{k-})/2i$. In addition, $\alpha(t)$ is coupled to these variables via number conservation and is given by

$$\alpha(t)^{2}=N/2-\sum_{k}\left[\langle\sigma_{kz}(t)\rangle\pm\frac{1}{2}\right]. \tag{5}$$

Assuming an initial vacuum state for the atoms, $|\mathrm{vac}\rangle$, we have $\langle\sigma_{kx}(0)\rangle=\langle\sigma_{ky}(0)\rangle=0$ and $\langle\sigma_{kz}(0)\rangle=\mp\frac{1}{2}$. For fermions (upper sign) Eqs. (4) are the Bloch equations describing the dynamics of a two-level system driven by the classical field $\alpha(t)$. For each $\boldsymbol{k}$ the motion is confined to the surface of the Bloch sphere defined by $\langle\sigma_{kx}\rangle^{2}+\langle\sigma_{ky}\rangle^{2}+\langle\sigma_{kz}\rangle^{2}=1/4$ and is an expression of the underlying Fermi statistics (see Fig. 1). On the other hand, for bosons (lower sign) the motion is confined to the surface of a one-sided three-dimensional (3D) hyperboloid defined by $\langle\sigma_{kz}\rangle^{2}-\langle\sigma_{kx}\rangle^{2}-\langle\sigma_{ky}\rangle^{2}=1/4$ and $\langle\sigma_{kz}\rangle\geqslant 1/2$, and the population for each $\boldsymbol{k}$ is unbounded (see Fig. 1).

It is instructive to consider the case when $\alpha(t)=\alpha_{0}$ is a constant. In this case, Eq. (4) can be easily solved for the above initial state to yield

$$\langle\sigma_{kz}(t)\rangle\pm\frac{1}{2}=\pm\frac{(g\alpha_{0})^{2}}{2\Omega_{k\pm}^{2}}[1-C_{k\pm}(t)], \tag{6}$$

where $\Omega_{k\pm}^{2}=(g\alpha_{0})^{2}\pm(\omega_{k}-\nu)^{2}$ and $C_{k+}(t)=\cos(2\Omega_{k+}t)$ and $C_{k-}(t)=\cosh(2\Omega_{k-}t)$. Again, the upper (lower) signs correspond to fermions (bosons). In the boson case this solution is unstable for $\alpha_{0}\neq 0$ as it leads to exponential growth at the rate $2\Omega_{k-}$ for all $\Omega_{k-}^{2}>0$. However, in the fermion case, the atom population is oscillatory about the Lorentzian-shaped mean value: $(g\alpha_{0})^{2}/2\Omega_{k+}^{2}$ and, in fact, we can find a self-consistent solution for α_{0} by substituting Eq. (6) back into Eq. (5) and assuming the oscillations eventually dephase for different ω_{k} (this procedure is compared to the numerical solution in Fig. 3). This solution is only valid when there is very little molecular decay, but it does indicate that a consistent solution can be found where the molecule population

does not completely decay away, and the numerical results presented below confirm this.

For illustrative purposes, we now confine our analysis to the case where ν is large so that the density of states is approximately flat across the region into which the molecules tend to decay. In this case, any deviation from exponential decay can be directly attributed to the quantum statistics of the atoms rather than any structure in the density of states (cf. [34]). Taking the continuum limit of Eq. (5) and evaluating the density of states at ν we can write

$$\alpha(t)^2 \approx N/2 - \rho(\nu)\int_{-\infty}^{\infty} d\delta\left[\langle\sigma_z(\delta,t)\rangle \pm \frac{1}{2}\right], \tag{7}$$

where $\rho(\nu)=V\sqrt{\nu/2}(m/\hbar)^{3/2}/\pi^2$ is the density of states at ν for a uniform 3D box of volume V. Since the equation of motion for the σ's only depends on $\delta=\omega_{\boldsymbol{k}}-\nu$ we have parametrized them by δ instead of $\boldsymbol{k}$. As discussed above, in the regime where there are ample states available to the atoms, the molecules decay exponentially and the atoms populate the frequencies δ with a Lorentzian distribution of width $\sim g^2\rho(\nu)$. A measure of the number of available states is therefore given by this width multiplied by the density of states: $\rho(\nu)$. Motivated by this, we introduce the dimensionless quantity $\Gamma=N/[g\rho(\nu)]^2$, such that the exponential-decay regime is given by $\Gamma \ll 1$. It follows that when this condition does *not* hold, we expect the behavior of the dissociation process to be altered by the quantum statistics of the atoms.

In Figs. 2 and 3 we have plotted the results of a numerical solution of Eqs. (4) and (5) for $\Gamma=4$ and $\Gamma=100$, respectively. Already for $\Gamma=4$ these plots show a marked deviation from the usual exponential decay. In particular, the molecular population in the fermion case does not decay to zero. For $\Gamma=100$ we see the accelerated decay that occurs due to bosonic stimulation in the boson case, whereas for the fermion case, a large population remains in the molecular state.

The behavior of the fermionic atoms can be qualitatively understood as follows: The molecular population initially undergoes a rapid decay into pairs of fermions. However, if $\Gamma \gtrsim 1$, the states close to resonance, $\delta \approx 0$, become filled and begin to undergo coherent association-dissociation oscillations, effectively halting the molecular decay. After a few oscillations the molecular population settles into a quasistationary state, α_0^2 (which we have only been able to determine numerically), leaving the spin vectors $\boldsymbol{S}(\delta)$ precessing about the effective "field" $\boldsymbol{B}(\delta)=[2g\alpha_0,0,2\delta]^t$, i.e., $\dot{\boldsymbol{S}}(\delta)=\boldsymbol{B}(\delta)\times\boldsymbol{S}(\delta)$. This results in the fringes in the distribution over δ (see insets in Figs. 2 and 3) which become more dense with time. The oscillation in the atomic population associated with this precession reacts back on the molecular field leading to an amplitude modulation of the molecular population which damps slowly due to the dephasing of the spins with different δ. In the final state the spins are completely dephased and no net dissociation or association can occur. This behavior is reminiscent of the processes of optical nutation, where an intense laser pulse excites an inhomogeneous media of two-level atoms [36]. Unlike the soliton-like oscillations predicted in Refs. [20], the oscillations described here are a transient phenomena, but have the advantage that they can be created in a straight-forward manner, experimentally. The incomplete dissociation for the fermion case was also found in Ref. [37] using a stochastic wavefunction approach.

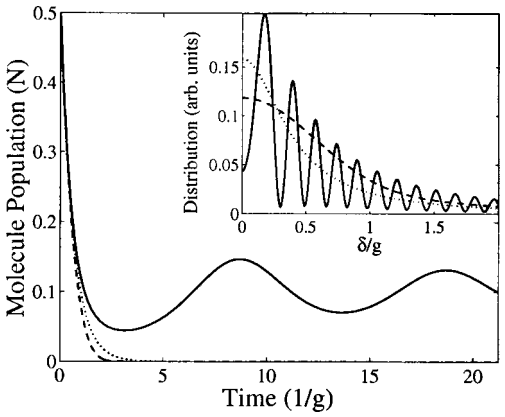

FIG. 2. Molecular population as a function of time for $\Gamma=4$ in the case where the decay products are pairs of bosons (dashed line) and pairs of fermions (solid line). For comparison the usual exponential decay at the rate $\gamma=\pi\rho(\nu)g^2$ is shown (dotted line). Inset: normalized population distribution of atoms over $\delta=\omega_{\boldsymbol{k}}-\nu$ at the final time $tg=21.2$ for bosons (dashed line) and fermions (solid line). The dotted line is a Lorentzian of width $\gamma/4$. Only the $\delta \geqslant 0$ case is shown as the distribution is symmetric about $\delta=0$.

Due to the assumption that the molecular condensate can be described by a mean field the atom pairs with different $\boldsymbol{k}$ are uncoupled and the Hilbert space of the system can be written as the tensor product $\mathcal{H}=\Pi_{\boldsymbol{k}}\otimes\mathcal{H}_{\boldsymbol{k}}$, where $\mathcal{H}_{\boldsymbol{k}}$ is the Hilbert space of the pair $(\boldsymbol{k}\uparrow,-\boldsymbol{k}\downarrow)$. We can write the unitary evolution operator as

$$U_{\boldsymbol{k}}(t)=\exp\{r_{\boldsymbol{k}}(t)[e^{i\phi_{\boldsymbol{k}}(t)}b_{\boldsymbol{k}\uparrow}^{\dagger}b_{-\boldsymbol{k}\downarrow}^{\dagger}-e^{-i\phi_{\boldsymbol{k}}(t)}b_{-\boldsymbol{k}\downarrow}b_{\boldsymbol{k}\uparrow}]\}, \tag{8}$$

where $r_{\boldsymbol{k}}(t)$ and $\phi_{\boldsymbol{k}}(t)$ are real time-dependent functions and are completely specified by the expectation values $\langle\sigma_{\boldsymbol{k}s}(t)\rangle$. Therefore, solving the Eqs. (4) enables us to determine not only the expectation values $\langle\sigma_{\boldsymbol{k}s}(t)\rangle$, but also the full quan-

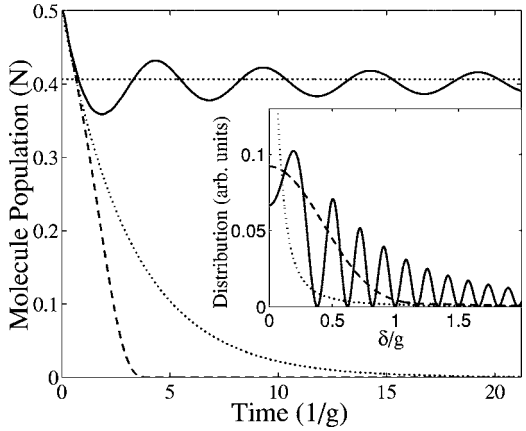

FIG. 3. Same as Fig. 2 but with $\Gamma=100$. The horizontal dotted line shows the steady-state molecular population determined by a self-consistent solution using Eqs. (5) and (6) (see text) which is a good fit in this case.

tum state of the atoms. In the case of bosons, Eq. (8) is analogous to the generator of a two-photon squeezed vacuum state [30]. In the case of fermions, when acting on the vacuum, this evolution operator yields the state

$$|\psi_{\boldsymbol{k}}(t)\rangle = [\cos r_{\boldsymbol{k}}(t) + b^{\dagger}_{\boldsymbol{k}\uparrow} b^{\dagger}_{-\boldsymbol{k}\downarrow} e^{i\phi_{\boldsymbol{k}}(t)} \sin r_{\boldsymbol{k}}(t)]|\mathrm{vac}\rangle, \quad (9)$$

which has obvious similarities to the BCS state.

For short times we expect that the dynamics will be dominated by the $\boldsymbol{q}=0$ condensate of molecules. However, the $\boldsymbol{q}\neq 0$ molecular modes, collisions and finite temperatures that have been neglected from this treatment are expected to lead to a slow redistribution of the atoms and a decay of the correlations [38]. Determining the relaxation dynamics to the new equilibrium state is beyond the scope of the present work, however, we can understand some aspects of the relaxation dynamics by adding phenomenological phase damping terms to the equations of motion: $d\langle\sigma_{\boldsymbol{k}j}\rangle/dt|_{\mathrm{relax}} = -\langle\sigma_{\boldsymbol{k}j}\rangle/T_2'$, where $j=x,y$. This relaxation process leads to a loss of the atom-molecule coherence ($\langle\sigma_{\boldsymbol{k}-}\rangle \to 0$) and will, in turn, destroy the dynamic equilibrium reached in the absence of damping. This intuition is confirmed by numerical simulations which show that for finite T_2' the molecular population always decays to zero and clearly demonstrates the key role played by the atom-molecule coherence, in combination with Pauli blocking, in the incomplete molecular dissociation in the fermion case.

Due to their narrow dissociation linewidth, narrow Feshbach resonances are promising systems to observe the effects described here and have the advantage that a magnetic field can be quickly tuned across the resonance into the dissociation regime. In fact, the dissociation scheme considered here has recently been used to measure the width of a number of Feshbach resonances of ^{87}Rb [39]. For the 912 G resonance with a width of $\Delta B=1.3$ mG, a detuning of $\hbar\nu=k_B\times 1$ μK yields a $\Gamma>3$ for typical densities ($n\sim 10^{13}$ cm^{-3}). Similarly, for the same parameters, the narrow 543 G resonance of ^{6}Li which has a width of $\Delta B=0.23$ G [2], yields a $\Gamma>12$, demonstrating that the regime where quantum statistics play a role are well within the reach of current experiments. Here we have assumed a uniform 3D system but note that the effects of quantum statistics can be significantly enhanced in systems of reduced dimensionality [13], or in the presence of trapping potentials, due to the reduction in the density of states [40].

In summary, we have studied the effects of quantum statistics on the dissociation dynamics of a condensate of diatomic molecules formed either by two bosonic or fermionic atoms. In the former case, the dissociation rate is Bose-enhanced; while for the latter, Pauli-blocking in combination with the coherence formed between the molecules and atom pairs lead to a dynamic equilibrium between the molecule and atom populations. Finally, we want to point out that we have used a method borrowed from quantum optics, which can serve as a powerful tool to treat other problems in the coupled atom-molecule system, such as the BEC-BCS crossover.

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