Dissociation dynamics of a Bose-Einstein condensate of molecules

Michael W. Jack and Han Pu
Department of Physics and Astronomy and Rice Quantum Institute, Rice University, Houston, Texas 77251
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An unstable condensate of diatomic molecules will coherently dissociate 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.

The ability to create quantum degenerate molecules composed of fermionic [1, 2, 3] or bosonic [4, 5] atoms by tuning a molecular level into resonance with the atomic states via a Feshbach resonance [6, 7, 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 ↔ 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, 13, 14, 15]. The case of diatomic bosonic molecules coupled to fermionic atoms (b ↔ ff) has also received a lot of attention lately due to the possibility of realizing a BEC-BCS crossover [16, 17, 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, 21, 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 non-equilibrium behavior [20, 22, 23, 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 [24, 25, 26, 27]. This highly non-equilibrium, 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., ν < 0 → ν > 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 molecular 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]. Here we present a unified treatment of the dissociation dynamics of a molecular condensate which includes both the boson (b ↔ bb) and fermion (b ↔ 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

\[
H = \hbar \omega_{0} a_{0}^{\dagger} a_{0} + \hbar \sum_{\mathbf{k}} \omega_{\mathbf{k}} (b_{\mathbf{k}\uparrow} b_{\mathbf{k}\downarrow} + b_{\mathbf{k}\downarrow}^{\dagger} b_{\mathbf{k}\uparrow}^{\dagger}) + hg \sum_{\mathbf{k}} \left( a_{\mathbf{k}\downarrow}^{\dagger} b_{-\mathbf{k}\downarrow}^{\dagger} b_{\mathbf{k}\uparrow} + a_{\mathbf{k}\uparrow} b_{\mathbf{k}\downarrow}^{\dagger} b_{-\mathbf{k}\downarrow}^{\dagger} \right),
\]

where \( a_{0} \) is the bosonic molecular mode and \( b_{\mathbf{k}\uparrow} \) and \( b_{\mathbf{k}\downarrow} \) are the annihilation operators of the atoms and either satisfy bosonic or fermionic commutation relations. \( \omega_{\mathbf{k}} = \hbar k^{2} / 2m \) 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, \( N = 2a_{0}^{\dagger} a_{0} + \sum_{\mathbf{k}} (n_{\mathbf{k}\uparrow} + n_{\mathbf{k}\downarrow}) \) is conserved, where \( n_{\mathbf{k}\uparrow} = b_{\mathbf{k}\uparrow}^{\dagger} b_{\mathbf{k}\uparrow} \) is the number operator for the atoms. As atoms of opposite spin and momentum are created and destroyed as pairs, the number difference, \( n_{\mathbf{k}\uparrow} - n_{-\mathbf{k}\downarrow} \), 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 [24] and references within). Quite recently, the pair correlations between fermionic atoms with opposite spin and momentum have been observed in the noise spectrum of photodissociated cold molecules [30], following the theoretical proposal of Ref. [31]. 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 oscillations [11]. This coherence—characterized by a non-zero value of \( \langle a_{\mathbf{k}\downarrow}^{\dagger} b_{-\mathbf{k}\downarrow} b_{\mathbf{k}\uparrow} \rangle \)—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 Hamiltonian 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 \[21, 21, 22\]. A similar mapping can be made for boson pairs and we treat the two cases in parallel. We define new operators by: \( \sigma_{k-} = b_k b_k^\dagger \), \( \sigma_{k+} = b_k^\dagger b_k \) and \( \sigma_{kz} = \frac{1}{2} (n_{kz} + n_{kz} - 1) \). It is easy to check that these operators satisfy the commutation relations: \( [\sigma_{k+}, \sigma_{k'+-}] = 0 \), \( [\sigma_{kz}, \sigma_{k'z}] = \delta_{k,k'} \sigma_{kz} \), and

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

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 \( H \) in terms of these new operators we have (minus a constant):

\[
H = \hbar \sum_k \left[ 2 (\omega_k - \nu) \sigma_{kz} + g (a_{k0}^\dagger \sigma_{k-} + \sigma_{k+} a_{0}) \right], \tag{3}
\]

and \( N = 2a_{0}^\dagger a_{0} + \sum_k (2 \sigma_{kz} \pm 1) \). 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. 22 and references within).

In the special case where the population in each mode is small, \( (n_{kz} + n_{kz}) \ll 1 \), 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 \[23\]. Under the Born-Markov approximation, which holds for weak coupling and large \( \nu \) \[24\], 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 \( \langle a_{0}^\dagger b_{k-} b_{k+} \rangle = \alpha \langle b_{-k} b_{k}^\dagger \rangle \) and the equations of motion for the averages \( \langle \sigma_{kz} \rangle \) can be written as:

\[
\frac{dS_k}{dt} = \begin{pmatrix}
0 & -2(\omega_k - \nu) & 0 \\
2(\omega_k - \nu) & 0 & +2g(\alpha(t)) \\
0 & 2g(\alpha(t)) & 0
\end{pmatrix} S_k, \tag{4}
\]

where \( S_k = [\langle \sigma_{kz} \rangle, \langle \sigma_{ky} \rangle, \langle \sigma_{kz} \rangle]^T \) 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

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

Assuming an initial vacuum state for the atoms, \( \langle \psi_{kz} \rangle \), we have \( \langle \sigma_{kz} \rangle = \langle \psi_{kz} \rangle = 0 \) and \( \langle \sigma_{kz} \rangle = \mp 2 \). 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 \( k \) 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) hyperbola defined by \( \langle \sigma_{kx} \rangle^2 - \langle \sigma_{ky} \rangle^2 = \langle \sigma_{kz} \rangle^2 = 1/4 \) and \( \langle \sigma_{kz} \rangle \geq 1/2 \), and the population for each \( k \) is unbounded (see Fig. 1).

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

\[
\langle \sigma_{kz}(t) \rangle = \pm \frac{1}{2} \frac{(g a_{0})^2}{2 \Omega_k^2} \left[ 1 - C_{k=\pm}(t) \right], \tag{6}
\]

where \( \Omega_k^2 = (g a_{0})^2 (\omega_k - \nu)^2 \) 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 \( a_{0} \neq 0 \) 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: \( (g a_{0})^2 / 2 \Omega_k^2 \) and, in fact, we can find a self-consistent solution for \( a_{0} \) by substituting Eq. 8 back into Eq. 4 and assuming the oscillations eventually dephase for different \( \omega_k \).
(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 \( \nu \) 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 (c.f. Ref. [32]). Taking the continuum limit of Eq. (4) and evaluating the density of states at \( \nu \)

\[
\alpha(t)^2 \approx N/2 - \rho(\nu) \int_{-\infty}^{\infty} d\delta \langle \sigma_2(\delta, t) \rangle \pm \frac{1}{2},
\]

where \( \rho(\nu) = V \sqrt{\nu/2} (m/\hbar)^{3/2}/\pi^2 \) is the density of states at \( \nu \) for a uniform 3D box of volume \( V \). Since the equation of motion for the \( \sigma \)'s only depends on \( \delta = \omega_k - \nu \) we have parameterized them by \( \delta \) instead of \( k \). 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 \( \delta \) with a Lorentzian distribution of width \( \sim \gamma \rho(\nu) \). 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 Figure 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 \( \Gamma \gg 1 \), the states close to resonance, \( \delta = 0 \), 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 quasi-stationary state, \( \alpha_\delta^2 \) (which we have only been able to determine numerically), leaving the spin vectors \( \vec{S}(\delta) \) precessing about the effective “field” \( \vec{B}(\delta) = [2g\alpha_\delta, 0, 2\delta] \), i.e. \( \dot{\vec{S}}(\delta) = \vec{B}(\delta) \times \vec{S}(\delta) \). This results in the fringes in the distribution over \( \delta \) (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 \( \delta \). 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 mutual annihilation of an intense laser pulse excites an inhomogeneous media of two-level atoms [35]. Unlike linear oscillations predicted in Refs. [20], the oscillations described here are a transient phenomenon, but have the advantage that they can be created in a straightforward manner, experimentally. The incomplete dissociation for the fermion case was also found in Ref. [36] using a stochastic wavefunction approach.

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

\[
U_k(t) = \exp \left\{ r_k(t) \left[ e^{i\phi_k(t)} b_{k\uparrow} b_{-k\downarrow} - e^{-i\phi_k(t)} b_{-k\uparrow} b_{k\downarrow} \right] \right\},
\]

where \( r_k(t) \) and \( \phi_k(t) \) are real time-dependent functions and are completely specified by the expectation values \( \langle \sigma_{k\uparrow}(t) \rangle \). Therefore, solving the Eqs. (4) enables us to determine not only the expectation values \( \langle \sigma_{k\uparrow}(t) \rangle \), but also the full quantum state of the atoms. In the case of bosons, Eq. (5) is analogous to the generator of a two-photon squeezed vacuum state [20]. In the case of fermions, when acting on the vacuum, this evolution op-
in turn, destroy the dynamic equilibrium reached in 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 turned 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 [38]. For the 912 G resonance with a width of $\Delta B = 1.3$ mG, a detuning of $\hbar \nu = k_B \times 1 \mu K$ 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 [23].

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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