1 Role of Molecular Dissociation in Feshbach-Interacting $^{85}$Rb Condensates

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Abstract

Recent Feshbach-resonance experiments with $^{85}$Rb Bose-Einstein condensates have led to a host of unexplained results: dramatic losses of condensate atoms for an across-resonance sweep of the magnetic field, a collapsing condensate with a burst of atoms emanating from the remnant condensate, increased losses for decreasing interaction times—until very short times are reached, and coherent oscillations between remnant and burst atoms. In particular, the amplitude of the remnant-burst oscillations, and the corresponding missing atoms, have prompted speculation as to the formation of a molecular condensate. Using a minimal mean-field model, we find that rogue dissociation, molecular dissociation to noncondensate atom pairs, is qualitatively implicated as the physical mechanism responsible for these observations, although very little molecular condensate is formed. Refining the model provides excellent quantitative agreement with the experimental remnant-burst oscillations, and the fraction of molecular condensate accounts almost entirely for the measured atom loss.

Introduction

The process known as the Feshbach resonance [1] occurs when two ultracold atoms collide in the presence of a magnetic field, whereby a spin flip of one atom can induce the pair to jump from the two-atom continuum to a quasibound molecular state. If the initial atoms are Bose condensed [2], the so-formed molecules will also comprise a Bose-Einstein condensate (BEC) [3]. Since the Feshbach resonance is mathematically identical to photoassociation [4, 5, 6], the process that occurs when two ultracold atoms form a molecule by absorbing a photon, insight gathered in either case is applicable to the other. In particular, the most recent results from photoassociation predict that rogue dissociation, molecular dissociation to noncondensate atom pairs, imposes a maximum achievable rate on atom-molecule conversion, as well as the possibility of coherent Rabi oscillations between the BEC and dissociated atom pairs [7].

Initial Feshbach resonance experiments in $^{85}$Rb [8] were motivated by the possibility [3] of enabling Bose condensation by tuning the natively negative atom-atom scattering length into the positive regime. As part of the experiment that achieved condensation [10], the magnetic field was swept across the Feshbach resonance, resulting in heavy condensate losses ($\sim 80\%$...
for the slowest sweep rates). Additional experiments led to the observation of collapsing condensates, an event characterized by bursts of atoms emanating from a remnant BEC, and coined "bosem nova" for the analogy with a supernova explosion [11]. More recently, experiments with pulsed magnetic fields that come close to, but do not cross, the Feshbach-resonance have revealed a striking increase in condensate loss for a decrease in the interaction time—until reaching very short times [12]. Finally, double-pulse results indicate large amplitude (∼25%) remnant-burst oscillations, with the missing atoms (∼10%) prompting speculation on the formation of a molecular condensate [13].

In the mean time, work on Feshbach-stimulated photoproduction of stable molecular condensates indicates that rogue dissociation dominates atom-molecule conversion for the above 85Rb Feshbach resonance, meaning that the production of a significant fraction of molecular BEC is not to be expected [14]. Additionally, we find intriguing the assertions of a breakdown of mean-field theory in the face of large resonance-induced scattering lengths [10, 12], especially given that theory actually faults the effective all-atom description [15]. We have therefore tested a mean-field model of coherent atom-molecule conversion against the salient features of the JILA experiments [16], and the subsequent understanding is presented in this Contribution (for related work, see Refs. [17, 18]).

Mean-field theory and its validity

The mathematical equivalence of the Feshbach-resonance [3] and photoassociation [4, 5] lies in both processes being described, in terms of second quantization, as destroying two atoms and creating a molecule. We therefore model a quantum degenerate gas of atoms coupled via a Feshbach resonance to a condensate of quasibound molecules based on Refs. [5, 6, 7]. The initial atoms are denoted by the boson field \( \phi(r, t) \), and the quasibound molecules by the field \( \psi(r, t) \). The Hamiltonian density for this system is

\[
\frac{\mathcal{H}}{\hbar} = \frac{1}{2} \left( \frac{\hbar \nabla^2}{2m} \right) \phi + \frac{1}{4} \left( \frac{\hbar \nabla^2}{4m} + \delta_0 \right) \psi \nonumber \\
- \frac{\Omega}{2\sqrt{\rho}} \left[ \psi^\dagger \phi \phi^\dagger \psi + \frac{2\pi \hbar a}{m} \phi^\dagger \phi \phi \right],
\] (1.1)

where

\[
\Omega = \lim_{\epsilon \to 0} \sqrt{\frac{2\pi \hbar^3/2 \rho \Gamma(\epsilon)}{\mu^{3/2} \epsilon}},
\] (1.2)

where \( m = 2\mu \) is the mass of an atom, \( \hbar \delta_0 \) is the energy difference between a molecule and two atoms, \( a \) is the off-resonant s-wave scattering length, \( \rho \) is an invariant density equal to the sum of atom density and twice the molecular density, and \( \Gamma(\epsilon) \) is the dissociation rate for a molecule with the energy \( \hbar \epsilon \) above the threshold of the Feshbach resonance.

To address the validity of mean-field theory for near-resonant systems, we find the Heisenberg equation of motion for the molecular operator and solve it in the adiabatic limit: \( \psi = [\Omega/(2\delta_0\sqrt{\rho})] \phi \phi \). Plugging this back into Eq. (1.1), gives

\[
\frac{\mathcal{H}}{\hbar} = \frac{2\pi \hbar a_{\text{eff}}}{m} \phi^\dagger \phi \phi; \quad a_{\text{eff}} = a \left[ 1 - \frac{m\Omega^2}{4\pi \hbar \delta_0 \rho a} \right].
\] (1.3)
The detuning can of course written in terms of the magnetic field by introducing the difference in the magnetic moments of the resonant states $\Delta \mu$: $\hbar \delta_0 = \Delta \mu (B - B_0)$. Similarly, the condensate coupling can be approximated as $\Omega = [2 \pi \rho |a| \mu ma \Delta R/m]^{1/2}$. Substituting these expressions leads immediately to the standard way of writing the scattering length around the Feshbach resonance:

$$a_{\text{eff}} = a \left(1 - \frac{\Delta B}{B - B_0}\right). \quad (1.4)$$

The effective scattering length (1.4), was obtained by adiabatically eliminating the molecular field, i.e., by assuming $\delta_0 \gg 2 \pi \hbar |a|/m, \Omega$. Since the adiabatic approximation fails for $\delta_0 \to 0$ ($B \to B_0$), it is therefore the effective all-atom description that actually breaks down when resonance is encountered [15], not mean-field theory. The validity of the mean field approximation is determined by $\rho |a|^3 \ll 1$, which holds independent of the value of the magnetic field. Including the molecular condensate dynamics provides a complete mean-field description of the near-resonant system.

That said, we expect the mean-field equations arising from the Hamiltonian (1.1) to suitably model the salient features of the JILA experiments. Switching to momentum space, only zero-momentum atomic and molecular condensate modes are retained, represented by the respective $c$-number amplitudes $\alpha$ and $\beta$. We also take into account correlated pairs of noncondensate atoms using a complex amplitude $C(\epsilon)$, which represent pairs of noncondensate atoms in the manner of the Heisenberg picture expectation value $\langle a_p a_{-p}\rangle$, with $\hbar \epsilon$ being the relative energy of the atoms. The normalization of our mean fields is such that $|\alpha|^2 + |\beta|^2 + \int d\epsilon |C(\epsilon)|^2 = 1$. We work from the Heisenberg equation of motion of the boson operators under the simplifying assumption that the noncondensate atoms pairs are only allowed to couple back to the molecular condensate, ignoring the possibility that noncondensate atoms associate to make noncondensate molecules. This neglect is justified to the extent that Bose enhancement favors transitions back to the molecular condensate. The final mean-field equations are [7]

$$i \dot{\alpha} = -\frac{\Omega}{\sqrt{2}} \alpha \beta,$$  
$$i \dot{\beta} = \delta_0 \beta - \frac{\Omega}{\sqrt{2}} \alpha \alpha - \frac{\xi}{\sqrt{2\pi}} \int d\epsilon \sqrt{\epsilon} C(\epsilon),$$  
$$i \dot{C}(\epsilon) = \epsilon C(\epsilon) - \frac{\xi}{\sqrt{2\pi}} \sqrt{\epsilon} \beta.$$  

(1.5)

(1.6)

(1.7)

The analog of the Rabi frequency for the rogue modes $\xi$ is inferred using Fermi Golden rule, which gives the dissociation rate for a positive-energy molecule as $\Gamma(\epsilon) = \sqrt{\epsilon} \xi^2$.

Next the problem is reformulated in terms of two key parameters with the dimension of frequency. The density-dependent frequency $\omega_\rho = \hbar \rho^{2/3}/m$, has been identified before, along with the operational significance that, once $\Omega \gtrsim \omega_\rho$, rogue dissociation is expected to be a dominant factor in the dynamics [5, 6, 7]. Here it is convenient to define another primary parameter with the dimension of frequency. Considering on-shell dissociation of molecules to atoms with the relative energy $\epsilon$, the Wigner threshold law delivers a dissociation rate $\Gamma(\epsilon)$ such that $\Gamma(\epsilon)/\sqrt{\epsilon}$ converges to a finite limit for $\epsilon \to 0$; hence, we define $4 \Xi = \dots$
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$(\lim_{\epsilon \to 0} \Gamma(\epsilon)/\sqrt{\epsilon})^2$, which indeed has the dimension of frequency. A combination of the preceding equations gives the parameters in the mean-field equations as

$$\Omega = 2^{3/2} \sqrt{\pi \Xi^{1/4}} \omega^{3/4}_b, \quad \xi = \sqrt{2} \Xi^{1/4}.$$  

(1.8)

Lastly, when the coupling to the continuum of noncondensate atom pairs is included, the continuum shifts the molecular state \cite{[19]}. We have, of course, taken the dominant state pushing, and the related renormalization effects, into account in our calculations \cite{[16, 20]}. Suffice it to say that we choose the bare detuning $\delta_0$ so that the renormalized detuning attains the desired value; hereafter, we use the symbol $\delta = \Delta \mu (B - B_0)/\hbar$ for the renormalized detuning, which is the parameter that is varied experimentally by changing the laser frequency in photoassociation, or the magnetic field in the Feshbach resonance. The position of the Feshbach resonance is $B_0 = 154.9$ G, and the difference in magnetic moments between bound molecules and free atom pairs, $\Delta \mu \approx 2 \mu_B$ (where $\mu_B$ is the Bohr magneton), is borrowed from $^{87}$Rb \cite{[21]}. We have estimated $\Xi = 5.29 \times 10^9$ s$^{-1}$, and thus $\xi = 381$ s$^{-1/4}$. Compared to the ensuing detunings $\delta$, the interactions energies between the atoms due to the background scattering length $a = 23.8$ nm are immaterial. We therefore ignore atom-atom interactions unrelated to the Feshbach resonance, as well as the (unknown) atom-molecule and molecule-molecule interactions.

Results

We begin with the experiments \cite{[10]} implementing a sweep of the magnetic field across the Feshbach resonance, which are of course a version of the Landau-Zener problem \cite{[5, 6, 22]}. Although a sweep of the detuning $\delta$ from above to below threshold at a rate slow compared to the condensate coupling $\Omega$ will move the system adiabatically from all atoms to all molecules, rogue dissociation will overtake coherent atom-molecule conversion when $\Omega > \omega_0^{3/4}$ \cite{[5, 6, 7]}. Nevermind that the JILA experiments sweep from below to above threshold, for a density $\rho = 1 \times 10^{12}$ cm$^{-3}$ the condensate coupling is $\Omega = 1.93 \times 10^5$ s$^{-1} \approx 250 \omega_b$, and so rogue dissociation should seriously dominate. This is indeed the case (see Fig. 1.1). Apparently, coherent conversion occurs not between atomic and molecular BEC, but between atomic BEC and dissociated atom pairs. Holding this thought, we conclude that mean-field theory indicates rogue dissociation as a primary sink of atoms in the Ref. \cite{[10]} sweeps across the resonance.

Next we consider the experiments \cite{[12]} for which nontrivial electromagnetic coil technology was developed to create trapezoidal magnetic field pulses that bring the system near--but not across-- resonance, hold for a given amount of time, and return to the original field value. Neglecting the burst, these remnant-focused experiments revealed a contradiction with the conventional understanding of condensate loss: rather than a loss that increased monotonically with increasing interaction time, the results indicated a loss that increased with decreasing interaction time, until very short times were reached. The present mean-field approach works similarly, as shown in Fig. 1.2. Our interpretation is that adiabaticity is again at play. At very short pulse durations, increasing interaction time leads to increasing condensate loss, as expected. In contrast, as the time dependence of the pulse gets slower, the system eventually follows the pulse adiabatically, and returns close to the initial condensate state when the pulse has passed.
Finally, we turn to the experiments [13] in which two trapezoidal pulses were applied to a $^{85}$Rb condensate, and the fraction of remnant and burst atoms measured for a variable between-pulse time and magnetic-field amplitude. These experiments revealed coherent remnant-burst oscillations with amplitudes of up to $\sim 25\%$. As it happens, we have recently predicted coherent oscillations between atoms and dissociated atom pairs in a rogue-dominated system, although we harbored doubts regarding any practical realization [7]. Casting these doubts aside, we consider a time dependent detuning (magnetic field) similar to Fig. 2 of Ref. [13] [Fig. 1.3(a)], and determine the fraction of remnant condensate atoms, noncondensate atoms, and molecules at the end of the pulse sequence as a function of the holding time between the two pulses [Fig. 1.3(b)]. Oscillations are seen with the amplitude of about 15% between condensate and noncondensate atoms at the frequency of the molecular state corresponding to the magnetic field during the holding period. The molecular fraction appears too small to account for the amplitude of the oscillations. In fact, what we termed molecular frequency is the characteristic frequency of a coherent superposition of molecules and noncondensate atom pair. Here the oscillations, directly comparable to Fig. 4(a) in Ref. [13], are Ramsey fringes [23] in the evolution between an atomic condensate and a molecular condensate dressed with noncondensate atom pairs.

Although our rogue-dissociation ideas provide a neat qualitative explanation for the three experiments we have discussed, in all fairness it must be noted that we have fallen short of a full quantitative agreement. We have therefore refined our renormalization techniques, extended our model to allow for Bose enhancement of the rogue modes, and included an average over a Gaussian distribution of densities [20]. So far, we have only applied this full model to the double-pulse experiments [13], the results of which are shown in Fig. 1.4. Not only do we find excellent agreement on the size of the experimental Ramsey fringes, but the fraction of molecular condensate ($\sim 5\%$) is now sufficient to explain the observed atom loss [8(3)\%].

Conclusions

In conclusion, we have demonstrated that a minimal mean-field model is sufficient to qualitatively explain a number of puzzling results in Feshbach-resonant systems [16]. Moreover, our refined model [20] gives near-perfect quantitative agreement with the double-pulse experiments [13], leaving little-to-no room for additional loss mechanisms. Collapsing-condensate physics is therefore understood as a matter of rogue dissociation, which leads to strong losses in the threshold neighborhood, decreased remnant fraction for decreasing interaction time—until very short times are reached, and coherent remnant-burst oscillations. Ironically, the Feshbach resonance has led to a regime dominated by rogue dissociation, which apparently tends to counteract the production of a molecular condensate.

\footnote{If an explanation in terms of below-threshold molecular dissociation seems a bit odd, consider that energy need not be conserved for transient processes where a time dependence is externally imposed on the system.}
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Figure 1.1: (a) Experimental [10] and theoretical (○) atom loss incurred in sweeping a $^{85}\text{Rb}$ BEC across the Feshbach resonance, where the magnetic field is swept in a linear fashion from $B_i = 162$ G to $B_f = 132$ G. In each numerical run, the fraction of molecular condensate is $\sim 10^{-6}$. (b) Results for $\ddot{B}^{-1} = 100$ μs/G are typical, and suggest that the system undergoes collective adiabatic following from BEC to dissociated atom pairs.
Figure 1.2: Theory of a magnetic field pulse applied to a $^{85}$Rb condensate for $\rho = 1.9 \times 10^{13}$ cm$^{-3}$ and $\Omega = 8.42 \times 10^5$ s$^{-1}$. (a) Remnant fraction versus detuning (magnetic field) rise time. (b-d) Results for a pulse with 150 $\mu$s rise time indicate adiabatic passage of BEC atoms to and from dissociated atom pairs. The minimum in panel (a), similar to Ref. [12], occurs at the onset of adiabaticity.
Figure 1.3: Simulation of the Ref. [13] experiments for a density $\rho = 5.4 \times 10^{13} \text{cm}^{-3}$ and $\Omega = 1.42 \times 10^6 \text{s}^{-1}$. (a) Time dependence of the detuning, and (b) the fraction of atoms in the remnant condensate (solid line), in noncondensate atoms pairs (dashed line) and in the molecular condensate (short-dashed line) after the pulse sequence as a function of the hold time $t_h$ between the two trapezoidal pulses. The frequency of the oscillations is compatible with our predictions for the molecular dissociation energy [16, 20], identifying these oscillations as Ramsey fringes in the transition between the atomic condensate and a molecular condensate dressed by dissociated atom pairs.
Figure 1.4: Full-model [20] simulation of the Ref. [13] experiments for a peak density $\rho_0 = 1.1 \times 10^{13}$ cm$^{-3}$. Fraction of atoms in the remnant condensate (solid line), in noncondensate atoms pairs (dashed line) and in the molecular condensate (dotted line) after the pulse sequence as a function of the hold time between the two trapezoidal pulses. The magnetic field pulse is similar to that shown in Fig. 1.3. Not only does the fringe amplitude agree quantitatively with the experimental observation, but the fraction of molecules formed is entirely consistent with the measured atom loss (see Fig. 6 of Ref. [13]).