Coherent Population Trapping in a Feshbach-Resonant $^{133}$Cs Condensate

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Recent experiments with Feshbach-resonant $^{133}$Cs Bose-Einstein condensates have led to unexplained molecule formation: a sudden switch of the magnetic field to its resonance value, followed by a finite hold time and another sudden switch to magnetic field values below threshold, converts about a third of the initial condensate atoms into molecules. Based on a model of coherent conversion between an atomic condensate, a molecular condensate, and magnetodissociated noncondensate atom pairs of equal and opposite momentum, we find that population trapping is strongly implicated as the physical mechanism responsible for molecule formation in switch experiments.

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Introduction. – Magnetoassociation creates a molecule from a pair of colliding atoms when one of the atoms spin flips in the presence of a magnetic field tuned near a Feshbach resonance \[1\]. Initial experiments with magnetoassociation of Bose-Einstein condensates (BECs) led to dramatic losses of condensate atoms in the neighborhood of resonance \[2\], a collapsing condensate with a burst of atoms emanating from the remnant condensate \[3\], increased losses for decreasing interaction times \[4\], and coherent oscillations between remnant and burst atoms \[5\]. Whereas atom-molecule coherence is both necessary and sufficient to explain these observations, the phase space densities are insufficient to warrant a claim to molecular condensate (MBEC) formation; hence, subsequent experiments focused on magnetic-field sweeps across the Feshbach resonance \[6\], which led to the unambiguous observation of BEC-MBEC conversion \[7\].

The backbone of these experiments is coherent evolution between an atomic condensate, a molecular condensate condensate, and magnetodissociated noncondensate atoms pairs of equal and opposite momentum \[8, 9\]. In particular, so-called rapid adiabatic passage arises because the ground state of the Feshbach system is all atoms far above the molecular-dissociation threshold and all molecules far below it, so that a slow sweep of the magnetic field from one extreme to the other converts atoms into diatomic molecules. The colloquial understanding is that a magnetic field sweep directed from below to above threshold converts an initial atomic condensate into noncondensate atom pairs of equal and opposite momentum \[8, 9\]. In particular, so-called rapid adiabatic passage arises because the ground state of the Feshbach system is all atoms far above the molecular-dissociation threshold and all molecules far below it, so that a slow sweep of the magnetic field from one extreme to the other converts atoms into diatomic molecules. The colloquial understanding is that a magnetic field sweep directed from below to above threshold converts an initial atomic condensate into noncondensate atom pairs of equal and opposite momentum \[8, 9\].

With this understanding in mind, magnetoassociation experiments with $^{133}$Cs condensates \[10\] have recently focused on creating molecules more efficiently \[11\]. For sweeps directed from above to below the threshold for magnetodissociation, the molecular conversion efficiency was found to saturate at about 10% for decreasing sweep rates, which is broadly similar to magnetic-field-sweep experiments with Rb \[11\] and Na \[7\]. Faced with this inefficiency, Mark et al. \[11\] decided to take another tack on making molecules: starting from well above threshold, they abruptly switch the magnetic field value to its resonance position, waited a given amount of time, and then abruptly switched the magnetic field value to a position well below threshold. Somewhat surprisingly, the result was a three-fold improvement in the molecular conversion efficiency. Moreover, the improved efficiency was independent of whether the system was initially above or below threshold, as long as the final switch was to a magnetic field value well below threshold. Obviously these observations defy an understanding in terms of adiabatic conversion of atoms into molecules \[8, 9\]. The purpose of this Letter is to demonstrate that molecular formation in the switch experiments can be explained as coherent population trapping–Rabi/Josephson oscillations that damp to a nonzero value–among the coupled atomic condensate, molecular condensate, and magnetodissociated atoms pairs.

Atom, Molecule, and Pair Model. – Early theories of collective magnetoassociation accounted only for the condensates, neglecting any and all noncondensate modes \[12\]. However, rogue \[8, 13\], or unwanted \[14\], transitions to noncondensate atom pairs can occur because magnetodissociation of a zero-momentum BEC molecule need not take the offspring atoms back to the zero-momentum atomic condensate, but may just as well end up creating two noncondensate atoms with equal-and-opposite momenta. Since the collective condensate coupling scales like the square root of the magnetic-field width of the Feshbach resonance and the magnetodissociation rate scales like the width itself, rogue dissociation

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is expected to play a dominant role in strong magnetoassociation. The prime indicator of rogue relevance is the density-dependent frequency \( \omega_p = \hbar \rho^{2/3}/m \). When the collective-enhanced atom-molecule coupling satisfies \( \Omega \gtrsim \omega_p \), the role of transitions to noncondensate atom pairs needs to be carefully considered in atom-molecule conversion.

The purpose of this section is to introduce our minimal-yet-realistic model of a joint atom-molecule condensate undergoing rogue dissociation to noncondensate atom pairs, in order to determine the mechanism behind the unexplained molecule conversion in the Innsbruck experiments. Hence, we assume that \( N \) atoms have Bose-condensed into the same one-particle state, e.g., a plane wave with wave vector \( \mathbf{k} = 0 \). Magnetoassociation then removes two atoms from this state \([1]\), creating a molecule in the molecular state \([2]\), which may then dissociate into the noncondensate pair state \( |\mathbf{k}, -\mathbf{k}\rangle \). In second quantized notation, boson annihilation operators for zero-momentum condensate atoms (molecules) and noncondensate atoms of momentum \( \mathbf{k} \) are denoted by \( a \) (\( b \)) and \( a_k \). The field-matter interactions that drive the atom-molecule transitions are characterized by the Rabi frequency \( \kappa \), the binding energy (detuning) of the molecule is denoted by \( \delta \); also, collisions between condensate particles are included with a coupling strength \( \lambda_{ij} \) (more on these quantities in a moment). The Hamiltonian for this system reads

\[
\frac{\hbar}{i} \frac{\partial}{\partial t} \psi = \mathcal{H}_0 + \mathcal{H}_{\text{int}},
\]

where \( \epsilon_k = \hbar k^2/2m \) is the energy of a noncondensate pair, and \( f_k \) describes the wavenumber (energy) dependence of the noncondensate coupling. Collisions with noncondensate atoms are neglected for simplicity.

The corresponding mean-field theory is derived from the Heisenberg equations of motion:

\[
\begin{align}
iv a &= \left[ \Lambda_{11} |a|^2 + \Lambda_{12} |b|^2 \right] a - \Omega a^* b, \\
iv b &= \left[ \delta + \Lambda_{12} |a|^2 + \Lambda_{22} |b|^2 \right] b - \frac{1}{2} \left[ \Omega a^2 + \xi \int d\epsilon \sqrt{\epsilon} f(\epsilon) C(\epsilon) \right], \\
iv C &= \epsilon C(\epsilon) - \xi \sqrt{\epsilon} f(\epsilon) [1 + 2P(\epsilon)] b, \\
iv \dot{P} &= \frac{(2\pi \omega)^{3/2}}{\sqrt{\epsilon}} f(\epsilon) [b^* C(\epsilon) - C^*(\epsilon) b].
\end{align}
\]

In Eqs. (2), we have taken the continuum limit

\[
\frac{1}{N} \sum_k G_k = \int d\epsilon G(\epsilon),
\]

and we also have introduced the rogue and normal densities, \( C(\epsilon) = \sqrt{\epsilon} \langle a_k a_{-k} \rangle/(2\pi \omega^{3/4}) \) and \( P(\epsilon) = \langle a_k^* a_k \rangle \), as well as the rogue coupling \( \xi = \Omega/(2\pi \omega^{3/4}) \). The effects of collective enhancement have been included by scaling the mean-field amplitudes according to \( x \to x/\sqrt{N} \) (where \( x = a, b \)). The mean-field fractions \( |a|^2 \) and \( 2|b|^2 \) are then of the order of unity, and the couplings have been redefined as \( \Omega = \sqrt{N} \kappa \) and \( \lambda_{ij} = N \lambda_{ij} \).

Finally, the collective-enhanced couplings are given explicitly as \( \Omega = |2\pi \hbar p|a_{11}|1| \Delta_p \Delta_B / m_{11}^* |^{1/2} \) and \( \lambda_{ij} = 2\pi \hbar p a_{ij}^* / m_{ij}^* \) (\( i, j = 1, 2 \)), where \( a_{11}, a_{12}, a_{22} \) denotes the zero-field atom-atom (atom-molecule, molecule-molecule) scattering length, \( \Delta_p \) denotes the difference between the magnetic moments of the free atom pair and the bound molecule, \( \Delta_B \) denotes the magnetic-field width of the Feshbach resonance, and \( m_{ij}^* \) is the reduced mass of the \( ij \)-th pair; also, the magnetic field dependence of the binding energy is given as \( \delta = \text{sgn}[a_{11}] \Delta_B (B - B_0)/\hbar \), where \( B_0 \) is the magnetic-field position of resonance.

Before moving on, we discuss basic numerics. For \( \Omega = \Lambda_{ij} = P = 0 \) and \( \xi \neq 0 \), simple Fourier analysis of an initial bound molecular condensate \( |b(t = 0) = 1\rangle \) gives the below-threshold binding energy as the real and negative pole of \( \omega - \delta - \Sigma(\omega) + i\eta = 0 \), where \( \eta = 0^+ \) and the molecular self-energy is defined as

\[
\Sigma(\omega) = \frac{\epsilon^2}{4} \int d\epsilon f^2(\epsilon) \frac{\sqrt{\epsilon}}{\omega - \epsilon + i\eta}. \tag{4}
\]

The simplest energy dependence for the continuum is one that obeys the Wigner threshold law up to some abrupt cutoff: \( f^2(\epsilon) = \Theta(\epsilon_M - \epsilon) \). The detuning (binding energy) then picks up a term \( \Sigma(0) = \epsilon^2 \sqrt{\epsilon_M} \). In principle, the cutoff is infinite, and therefore so is the continuum shift of the molecular binding energy. To account for this divergence, one defines the so-called physical detuning \( \delta = \delta - \epsilon^2 \sqrt{\epsilon_M} \), which is finite by definition in the limit of an infinite cutoff. In practice, any numerical procedure employs a finite cutoff, and the finite shift is accounted for in exactly the same manner, and the intermediate detunings in Eqs. (2) are taken as physical (renormalized) detunings. The number of noncondensate quasicontinuum states and cutoff are chosen to deliver convergence and minimize numerical artifacts.

**Results for Sweep and Switch Experiments.**–To enable a discussion of the \(^{133}\text{Cs}\) sweep and switch experiments, we gather explicit parameters from Refs. [10, 11]: \( a_{11} = \)
200a_0, \Delta \rho = 0.93\mu_0, \Delta B = 2.1\,\text{mG}, \text{and } B_0 = 19.84\,\text{G} \text{ [where } a_0 (\mu_0) \text{ is the Bohr radius (magneton)]. Although exact values are unknown, we approximate the effect of atom-molecule and molecule-molecule collisions with } \Lambda_{12} = \Lambda_{22} = \Lambda_{11}; \text{ similarly, we account for collision-induced vibrational relaxation by borrowing an imaginary scattering length from Na (see, e.g., Yurowsky et al. [12]), so that } \Omega(\Lambda_{12}) = \Omega(\Lambda_{22}) = \rho \times 10^{-11} \,\text{s}^{-1}. \text{ Crucially, } \Omega/\omega_\rho \sim 2 \text{ for } \rho \sim 10^{12} \,\text{cm}^{-3}, \text{ so that rogue dissociation to noncondensate modes must be treated carefully, despite the narrow resonance [13].}

In order to ensure a uniform magnetic field across the atomic sample, the combined optical-dipole and magnetic-field-levitation trap is turned off, leading to ballistic expansion over the course of the experiments [11]. Nevertheless, we simply assume a peak density consistent with \( \sim 10 \text{ ms of ballistic expansion, } \rho_0 = 1.1 \times 10^{12} \,\text{cm}^{-3} [11], \text{ and account for the initial inhomogeneity with a local density approximation. This overestimates the density and, in turn, the role of vibrational quenching; on the other hand, the magnetic-field range here is truncated, and the sweep rate decreased, which ultimately shortens the total sweep time and so underestimates relaxational losses. Of course, these two factors do not necessarily cancel each other out, and the ensuing results are only qualitative; the aim is not to quantitatively model the sweep experiments anyway, but to explain the improved efficiency of the switch experiments.}

In the sweep experiments [11], the magnetic field is initially tuned 0.5 G above the Feshbach resonance threshold, \( B_t = B_0 + 0.5 \,\text{G} \), which is ramped in a linear fashion, \( B(t) = B_0 - B_t \), to a final value 0.5 G below threshold, \( B_f = B_0 - 0.5 \,\text{G} \). For experimental ramps with \( \dot{B} \lesssim 10 \,\text{G/s} \) the total sweep time is \( \gtrsim 100 \,\text{ms} \), and the conversion efficiency saturates at ca. 10%. For numerical ease the B-field range here is truncated to \( B = B_0 \pm 20 \Delta B \), and we use \( \dot{B} = 1 \,\text{G/s} \) for a total sweep time of 84 ms. The results are shown in Fig. [11] The agreement is surprisingly good, all things considered. Also, as the noncondensate population begins to build, it is transferred into the molecular BEC; hence, creation of molecules instead of dissociated pairs is more about coherent conversion than a closing magnetodissociation channel (the effect is even more dramatic for slower sweeps, as per Ref. [14]).

The relevant frequency scale is \( \Omega \sim \Lambda_{13} \), so that adiabatic following requires \( \delta/\Omega \lesssim 1 \). For \( \rho_0 \) and \( \dot{B} \) as above, \( \delta/\Omega_\rho \approx 8 \); hence, the center of the cloud is still on the outside of the adiabatic regime, and the situation is worse on the edges of the cloud. Slower sweeps therefore lead to more molecules, but allow more time for vibrational relaxation, leading to a saturation of the molecular conversion efficiency. Nevertheless, it is only necessary to satisfy adiabaticity in the immediate vicinity resonance, so it should be worth trying to engineer a ramp that is “ultrafast” except in the region \( B \sim B_0 \pm \Delta B \). In this manner, adiabaticity could be satisfied in the vicinity of resonance, while minimizing the total sweep time and, thus, vibrational relaxation losses, yielding more MBEC.

Now we turn to the Ref. [11] switch experiments. Here the magnetic field is suddenly switched to \( B \sim B_0 \) in a time \( \sim 10 \text{ ms} \), held for a given amount of time, and then suddenly switched to a well-below-threshold value where the fraction of molecules is determined. Ballistic expansion is not really an issue when modeling the switch experiments: the switch occurs in a time \( \sim 10 \text{ ms} \), and the hold times are \( \sim 10 \text{ ms} \) themselves, so it is entirely reasonable to use \( \rho_0 = 1.1 \times 10^{12} \,\text{cm}^{-3} \) throughout; moreover, the switch time here is shortened, without changing the physics, to hasten the numerics. The results are shown in Fig. [2] Whereas not much happens during the switch period, during the hold period the system undergoes coherent Josephson/Rabi oscillations between the atomic condensate, the molecular condensate, and noncondensate atom pairs; however, while the couplings remain, the system subsequently freezes into a superposition immune to magnetodissociation. The sudden switch back then harvests the frozen population. The independence of the whether the initial B-field is above or below threshold is seen by comparing Figs. (a,b). More importantly, an intriguing feature of the experiments is an apparent step-wise threshold in the conversion efficiency at \( \sim 15 \text{ ms} \), beyond which the molecular efficiency is \( \sim 30\% \). Due to an inadvertent lag [13], the B-field value does not reach the vicinity of resonance as expected, so the system is still off resonance; along with the fact that trapped-population fraction is depends on the detuning, as per Figs. (c,d), the lag leads to an apparent threshold. Similarly, experimental fluctuations in the magnetic

![FIG. 1: Coherent conversion of a $^{133}\text{Cs}$ atomic condensate into a $^{133}\text{Cs}_2$ condensate by sweeping a magnetic field across a Feshbach resonance. Here $\dot{B} = 1 \,\text{G/s}$, making for a total sweep time of 84 ms, which is, evidently, plenty of time for vibrational relaxation to filch $\sim 5\%$ of the MBEC population.](image-url)
Conclusions.—Given that faster sweeps apparently convert fewer atoms into molecules, improving the molecular conversion efficiency by speeding up the sweep is truly a surprise. The plot thickens with the observation of a final $^{133}$Cs$_2$ fraction that is independent of whether the initial magnetic field is tuned above or below threshold (as long as the final switch is to below threshold), in contrast with previous experiments [6, 7]. Nevertheless, it is known from past work on photodissociation of a negative ion [17] that—given a shaped continuum—some of the population may stay permanently trapped in the bound state. Indeed, the same effect was predicted for strong photoassociation [13], and since the theory of a photoassociation and magnetooassociation resonance are formally equivalent, the answer is obvious in hindsight. And yet it is still surprising that, given all the possible decoherence avenues, coherent population trapping could survive. The results herein strongly suggest that it does, making coherent population trapping the leading candidate to explain how a fast switch could get the better of a slow sweep.

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