Zero-Temperature Theory of Collisionless Rapid Adiabatic Passage from a Fermi Degenerate Gas of Atoms to a Bose-Einstein Condensate of Molecules

Matt Mackie¹ and Olavi Dannenberg²

¹QUANTOP–Danish National Research Foundation Center for Quantum Optics, Department of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus C, Denmark
²Helsinki Institute of Physics, PL 64, FIN-00014 Helsingin yliopisto, Finland

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We theoretically examine a zero-temperature system of Fermi degenerate atoms coupled to bosonic molecules via collisionless rapid adiabatic passage across a Feshbach resonance, focusing on saturation of the molecular conversion efficiency at the lowest magnetic-field sweep rates. Borrowing a novel many-fermion Fock-state theory, we find that a proper model of the magnetic-field sweep can systematically remove saturation. We also debunk the common misconception that many-body effects are responsible for molecules existing above the two-body threshold.

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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]. Recently, ultracold [2, 3] and condensate [4] molecules have been created via magnetoassociation of a Fermi gas of atoms, in the course of efforts to create superfluid Cooper-paired atoms [5, 6] (see also Refs. [7]). The backbone of these experiments is rapid adiabatic passage: 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.

Finite-temperature mean-field theory of magnetoassociation of a Fermi gas of atoms leads to two types of instabilities against molecule formation. One is the thermodynamic instability of a Fermi sea against the formation of Cooper pairs [8], a trait of superconductors whose analog is passed on to Feshbach-resonant superfluids [9]. A thermodynamical instability occurs because pairing lowers the energy, and coupling to a reservoir with a low enough temperature leaves the system prone to pairing. The other is a dynamical instability, whereby the larger state space of the molecules, owing somewhat to Pauli blocking, leaves the atoms prone to spontaneous association [10]. The role that temperature plays in this process is an open question experimentally, as well as a matter of theoretical contention.

Physically [10], high temperature experimentally lessens the chance of an atom occupying an arbitrary level in the Fermi sea, the dynamical instability becomes less effective and the efficiency of even the slowest rapid adiabatic passage therefore saturates (c.f., Fig. 1). The mean-field theory behind this understanding agrees semi-quantitatively with experiments [2]: nevertheless, a recent zero-temperature Landau-Zener theory predicts that saturation is fundamental to the collisionless regime [11]. If temperature is not a limiting factor, then any zero-temperature model of collisionless rapid adiabatic passage should ultimately display saturation, e.g., a Fock-state approach similar to the theory of cooperative association of Bose-Fermi mixtures of atoms into Fermi molecules [12]. Unfortunately, computing power is presently sufficient for calculations with only about 20 atoms total at best, precluding any brute-force test of saturation. Here we apply a novel large-fermion-number theory [13] to demonstrate near-unit-efficient collisionless rapid adiabatic passage in the limit of zero temperature, thereby ruling out any fundamental ceiling to the molecular conversion, and bolstering Ref. [10] (and also Ref. [14]).

This development is outlined as follows. After briefly introducing the collisionless model, we focus on rapid adiabatic passage and confirm the reduced-space mapping [15] by comparison with exact few-particle results. Increasing the total particle number to 2×10², we then observe what appears to be saturation at about ~50%. However, including fluctuation effects in the rate at which the system is swept across the Feshbach resonance, we...
find that saturation can be systematically removed, and near-unit efficiency can be achieved for any particle number. Lastly, from the single pair results we also debunk the commonly held notion that many-body effects are responsible for the existence of molecules above the threshold for molecular dissociation.

Collisionless Gas Model.—We model an ideal two-component gas of fermionic atoms coupled by a Feshbach resonance to bosonic molecules. In the language of second-quantization, an atom of mass \( m \) and momentum \( \mathbf{k} \) is described by the annihilation operator \( a_{\mathbf{k}, \sigma} \), and a molecule of mass \( 2m \) and similar momentum is described by the annihilation operator \( b_{\mathbf{k}} \). All operators obey their (anti)commutation relations. The microscopic Hamiltonian for such a freely-ideal system is written

\[
\frac{\hbar}{\sigma} = \sum_{\mathbf{k}} \left[ (\varepsilon_k - \mu) a_{\mathbf{k}, \sigma}^\dagger a_{\mathbf{k}, \sigma} + \left( \frac{\hbar}{2} \varepsilon_k + \delta - \mu_{\text{mol}} \right) b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \right] + \kappa \sum_{\mathbf{k}, \mathbf{k}'} \left[ b_{\mathbf{k}+\mathbf{k}'}^\dagger a_{\mathbf{k}, \sigma} b_{\mathbf{k}', \sigma} + \text{H.c.} \right],
\]

(1)

where repeated greek indices imply a summation (\( \sigma = 1, 2 \)). The free-particle energy is \( \hbar \varepsilon_k = h^2 k^2 / 2m \), the atom (molecule) chemical potential is \( \mu_{\text{atom}} \), and the detuning \( \delta \) is a measure of the binding energy of the molecule (\( \delta > 0 \) is taken as above threshold), the model-independent atom-molecule coupling is \( \kappa \propto 1/\sqrt{V} \) with \( V \) the quantization volume.

We have already imposed the ideal conditions for atom-molecule conversion with \( \mu_1 = \mu_2 = \mu \). An appropriate unitary transformation then shuffles \( \mu \) into the definition of \( \mu_{\text{mol}} \) which, in turn, can be absorbed into the detuning and written off as an effectively dc bias (see also Refs. [12]). Since magnetoassociation usually occurs much faster than any trapped-particle motion, an explicit trap can be neglected along with the free-particle energies \( \varepsilon_k \). For the sake of simplicity, and to compare with Ref. [10], we neglect all molecular modes except the \( \mathbf{k} + \mathbf{k}' = 0 \) mode, \( b_{\mathbf{k}} \equiv b_0 \), so that

\[
\frac{\hbar}{\sigma} = \delta b_{\mathbf{k}}^\dagger b_0 + n \sum_{\mathbf{k}} \left[ b_{\mathbf{k},1}^\dagger a_{\mathbf{k},1} - b_{\mathbf{k},2}^\dagger a_{\mathbf{k},2} + \text{H.c.} \right].
\]

(2)

Absent losses, the total particle number is conserved, 

\[
2 \langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \rangle + \sum_{\mathbf{k}} \langle a_{\mathbf{k},1}^\dagger a_{\mathbf{k},1} \rangle = 2n + \sum_{\mathbf{k}} \nu_{\mathbf{k}, \sigma} = 2N,
\]

where \( n \) is the number of molecules, \( \nu_{\mathbf{k}, \sigma} = 0,1 \) is the number of atoms per mode (\( \mathbf{k} \)) per species (\( \sigma \)), and \( 2N \) is the total number of atoms were all the molecules to dissociate. For a fixed number of particles equal to the number of fermion modes, the Fock-state wavefunction is

\[
|\psi(t)\rangle = \sum_{m' = 0}^{N} \sum_{n_{1}, \ldots, n_{N}} C_{N-m', n_{1}, \ldots, n_{N}}(t) |N-m', n_{1}, \ldots, n_{N}\rangle
\]

(3)

The time dependence of the system is determined by the Schrödinger equation, \( i\hbar \partial_t |\psi\rangle = \mathcal{H} |\psi\rangle \), so that the Hamiltonian \( \mathcal{H} \) yields

\[
i\mathcal{C}_m = \left[ N - m \right] \delta \mathcal{C}_m + \kappa \left[ \sqrt{N - m} + 1 \right] D_{m-1} \mathcal{C}_{m-1} + \kappa \left[ \sqrt{N - m} D_{m+1} \mathcal{C}_{m+1} \right].
\]

(4)

Here \( \mathcal{C}_m(t) \equiv C_{N-m, n_{1}, \ldots, n_{N}}(t) \) is a column vector of all the amplitudes corresponding to the \( (N \choose m) \) possible arrangements of \( m \) atom pairs among the \( N \) available fermion modes, and \( D_{j} \) is an \( (N \choose j) \times (N \choose j) \) dimensional matrix that contains only unit and zero elements determined by \( \mathcal{C}_j \) and \( \mathcal{C}_j \). The problem with the system is that there are \( 2^N \) amplitudes, which limits most numerical experiments in rapid adiabatic passage to about \( N = 10 \) (see also Refs. [12]); however, by multiplying Eqs. (4) by the appropriate column vector \( \mathbf{u}_{m, N} \), any redundant amplitudes can be eliminated. The remaining \( N+1 \) amplitudes evolve in time according to

\[
i\mathcal{\alpha}_m = \left[ N - m \right] \delta \mathcal{\alpha}_m + \kappa \left[ \sqrt{m} (N - m + 1) \mathcal{\alpha}_{m-1} + \sqrt{m + 1} (N - m) \mathcal{\alpha}_{m+1} \right],
\]

(5)

where the sum of all \( (N \choose m) \) amplitudes with \( N - m \) molecules and \( m \) free atom pairs is defined as \( \sqrt{N} \mathcal{\alpha}_m = \mathbf{u}_{m, N} \mathcal{C}_m = \sum_{n_{1}, \ldots, n_{N}} C_{N-m, n_{1}, \ldots, n_{N}} \) (with \( \mathcal{\alpha}_m \) normalized to the number of permutations of \( m \) atoms in \( N \) states). Lastly we will need the molecular fraction \( |\beta|^2 = 2 \langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \rangle/(2N) = (1/N) \sum_{m=0}^{N} (N - m) |\mathcal{\alpha}_m|^2 \).

Rapid Adiabatic Passage.—Putting fluctuations momentarily aside, the relevant frequency scale is \( \Omega = \sqrt{\hbar K} \propto \sqrt{\rho} \equiv \sqrt{\langle n_{\mathbf{k}} a_{\mathbf{k}, \sigma}^\dagger a_{\mathbf{k}, \sigma} \rangle} \), where \( \sqrt{\rho} \) is the so-called collective enhancement factor. “Adiabatic” is therefore defined qualitatively as the detuning changing by an amount \( \Delta \) in a time \( 1/\Omega \), or \( |\delta| \lesssim \Omega^2 \). Modeling the time dependent detuning as \( \delta = -\xi \Omega^2 t \), sweeps with \( \xi \sim 1 \) should qualify as adiabatic. Off hand, Fig. (2a) confirms this intuition for \( N = 4 \); also, noting that the full results are shifted for clarity, the reduced system (4) indeed reproduces the full system (3). Making a more full use of the reduced-space theory, Fig. (4b) illustrates that the efficiency of rapid adiabatic passage in fact decreases for increasing particle number, saturating at about 50% for \( N = 10^2 \). Nevertheless, if we account for fluctuations, then the relevant frequency scale is \( \Omega/\ln N \). Now the detuning should change by \( \Omega/\ln N \) in a time \( (\Omega/\ln N)^{-1} \), suggesting the detuning-sweep model \( \delta (t) = -\xi (\Omega/\ln N)^2 t \). Indeed, Fig. (2c) shows that the \( N = 10^2 \) and \( N = 1 \) results agree nicely, and are absent any evident saturation.

We can also make a rough comparison with the zero-temperature limit in Fig. (1). Magnetic fields are converted into detunings according to \( \delta = \Delta_{\mu} (B - B_0) / \hbar \), where the difference in magnetic moments between the atom pair and a molecule is \( \Delta_{\mu} \), and \( B_0 \) is the magnetic-field position of resonance. For \( N = 10^5 \) atoms of \({}^{40}\text{K}\) in a typical trap the peak density \( \rho = 2 \times 10^{13} \text{cm}^{-3} \), so
that the coupling strength is $\Omega = 0.3 \times 2\pi$ MHz \cite{10}; the difference in magnetic moments is $\Delta \mu \approx 0.19 \mu_B$ \cite{10}, where $\mu_B$ is the Bohr magneton. The results of Fig. 4 are for $1/B = 400 \mu \text{G}$ \cite{10}, which corresponds to $\xi \approx (\ln N)^2 \Delta \mu / (\hbar \Omega^2) \approx 7.9$ for $N = 10^5$ atoms per species. Of course, even the reduced-space model \cite{13} cannot handle $N = 10^5$ atoms, but for $\xi \approx 7.9$ then $N = 10^2$ will actually underestimate the $N = 10^5$ results. Hence the already good agreement between Fig. 2(d) and Fig. 4 would actually improve if resources were available to manage the correct number of particles.

We pause briefly to justify the ideal gas model. The collisional interaction strength is roughly $\Lambda = 2\pi \hbar a_0 / m$, where $a_0$ is the off-resonant atomic $s$-wave scattering length. The $^{40}\text{K}$ scattering length is $a = 176 a_0$ \cite{21}, with $a_0$ the Bohr radius. For a typical density $\rho \approx 10^{13}$ cm$^{-3}$, the collisional coupling strength, in units of the atom-molecule coupling, is $|\Lambda|/\Omega \approx 10^{-3}$. Collisions should therefore be broadly negligible. In particular, a system of Fermi atoms coupled to Bose molecules is formally identical to a system of only bosons \cite{13}, and collisions are negligible for bosons under such conditions \cite{18}.

Also, it should be noted that, because we have chosen $\Omega$ as the frequency scale, the above results are automatically for a resonance (atom-molecule coupling) of arbitrary strength. However, the model \cite{15} is broadly equivalent to the two-mode model in coherent association of condensate \cite{13}, and it is well known that strong coupling can lead to dissociation to modes lying outside the two-mode system \cite{12,15}, so-called rogue dissociation. Nevertheless, if the sweep is directed from above to below threshold, then rogue dissociation is negligible and the two-mode model is a good approximation. Hence, the above results are expectedly reasonable to describe a sweep across an arbitrarily strong resonance.

Before closing, we turn to a related matter of principle: the nature of above-threshold molecules. Below threshold ($\delta < 0$), Fourier analysis delivers the binding energy, $\hbar \omega_B < 0$, of the Bose-condensed molecules \cite{11,19}: $\omega_B - \delta - \Sigma'_{B} + i \eta = 0$, where $\Sigma'_{B} = 0$ is the finite self-energy of the Bose molecules and $\eta = 0^+$. Tuning the system above the two-body threshold ($\delta > 0$) gives an imaginary $\omega_B$, and the bound state ceases to exist; nevertheless, Fig. 3 shows a large $N = 1$ molecular fraction. This result is not really a surprise, since the fraction of molecules must vary continuously from zero to unity across threshold. We conclude that any theory in which molecules abruptly cease to exist at threshold, while useful in their own right (e.g., for modeling binding energies \cite{10}), are not a good rule of thumb for predicting the existence of above-threshold molecules. Our interpretation is that, as usual in cooperative behavior, a macroscopic number of particles respond as a unit to a given external drive, thereby mimicking one- or two-body physics. At the least, this implies that many-body effects are sufficient but not necessary for the existence of above-threshold molecules. Moreover, we see in Fig. 3 that the above-threshold molecular fraction for $\delta/\Omega \approx 2$
is actually suppressed for the many-body case $N = 10^2$.

Of course, the idea of many-body stabilization of above-threshold molecules generally arises in the context of equilibrium thermodynamics, whereas the collisionless model describes non-equilibrium processes. The many-body suppression of the above-threshold molecular fraction may or may not carry over to the collisional regime (although we find elsewhere that, to a certain degree, it may [21]). However, we expect that the two-body equilibrium thermodynamics, whereas the collisionless model describes non-equilibrium processes. The many-body effects to explain the existence of above-threshold molecules.

**Conclusions.**–We have investigated saturation in collisionless rapid adiabatic passage from a two-component degenerate Fermi gas to a Bose-Einstein condensate of molecules. Saturation indeed arises, but can be systematically eliminated by introducing the timescale appropriate to cooperative interference effects. Physically, cooperative interference effects arise from adding up the various pathways coupling the states having $N - m$ molecules and $m$ dissociated atom pairs with the states having one more (less) molecules and one less (more) dissociated pair, and the timescale for $N$-particle interference turns out to be $\sim \ln N/\Omega$[13]. It then makes perfect sense that cooperative (near-unit-efficient and macroscopic) rapid adiabatic passage will only occur over a timescale that is commensurate with constructive interference. Next we saw that our zero-temperature model agrees semi-quantitatively with our mean-field model[10], indicating that temperature[10] and pair correlations[14] are—as of yet—the main obstacles to collisionless cooperative conversion to molecules with near-unit efficiency. Finally, whereas studies of Feshbach resonances for both fermions and bosons have implicated many-body effects in the existence of molecules above the two-body threshold for dissociation, we find that it is not necessary to invoke many-body effects to explain the existence of above-threshold molecules.

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