Shortcut to a Fermi-Degenerate Gas of Molecules via Cooperative Association

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We theoretically examine the creation of a Fermi-degenerate gas of molecules by considering a photoassociation or Feshbach resonance applied to a degenerate Bose-Fermi mixture of atoms. This problem raises an interest because, unlike bosons, fermions in general do not behave cooperatively, so that the collective conversion of, say, two million atoms into one million molecules is not to be expected. Nevertheless, we find that the coupled Fermi system displays collective Rabi-like oscillations and adiabatic passage between atoms and molecules, thereby mimicking Bose-Einstein statistics. Cooperative association of a degenerate mixture of Bose and Fermi gases could therefore serve as a shortcut to a degenerate gas of Fermi molecules.

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Laser cooling of molecules is difficult, if not impossible, to apply in practice. This bottleneck occurs because the removal of energy from the translational degrees of freedom is most often accompanied by excitation of ro-vibrational modes which, upon de-excitation, couple back into the translational motion in a manner that heats the system. A laser-cooling-based approach [1] to a Fermi-degenerate gas of molecules is therefore unlikely to succeed. Moreover, buffer-gas cooling [2] and Stark-deceleration [3] techniques, while no doubt of practical interest in their own right, currently view quantum degeneracy from the horizon.

One possible route to ultracold molecules is photoassociation [4], and the formation of nondegenerate Fermi molecules was indeed recently observed [5]. A shortcut to quantum degenerate molecules is in the early stages of development for Bose-Einstein condensates (BECs), so far delivering strongly enhanced molecule formation [6], precise measurements of the light-induced shift of the binding energy [7], and searches for a fundamental limit to the atom-molecule conversion rate [8,9]. The magnetic field counterpart to photoassociation is the Feshbach resonance [10]. Often seen as a tool for adjusting the scattering lengths, thereby enabling condensation in otherwise-incondensible systems [11,12], the latest studies of a BEC tuned near a Feshbach resonance [10] have shed light on the role of atom-molecule interactions in the transition to noncondensate-like behavior [13].

Motivated by these exciting experiments, we report that a degenerate gas of Fermi molecules could be formed by applying either a photoassociation [5–9] or Feshbach resonance [11–13] to a degenerate mixture of Bose and Fermi atoms [14–16]. The collective Fermi atom-molecule system is predicted to undergo Rabi-like oscillations, as well as adiabatic passage, from atoms to molecules, thereby mimicking cooperative behavior that was previously attributed to Bose statistics [17,18].

Photoassociation occurs when two atoms absorb a laser photon, thereby jumping from the free two-atom continuum to a bound molecular state. If the initial atoms form a Bose-Einstein condensate, then a photoassociation laser could conceivably be used to convert a BEC of atoms into a BEC of molecules [19–21]. Collective free-bound photoassociation is theoretically identical to magnetoassociation, whereby a molecular condensate can be created when one atom from a condensed pair spin flips in the presence of a magnetic field tuned near a Feshbach resonance [22–24]. Intuition developed in one instance is therefore generally applicable to the other, and we will often refer simply to collective association.

Analogous with coherent optical transients in few level atomic systems [25], photoassociation of a BEC has been predicted to induce Rabi-like oscillations between atomic and molecular condensates [19–21], whereby an entire gas of, say, two million Bose-condensed atoms are collectively converted into a million molecules that are, in turn, collectively converted back into (roughly) two million atoms, ad infinitum. Another interesting possibility arises because the ground state of the system is all atoms for large positive detunings (far below threshold) and all molecules for large negative detunings (far above threshold), so that a slow sweep of the laser detuning from one extreme to the other will collectively convert a BEC of atoms into a BEC of molecules [19]. Incidentally, it was a particular combination of these two concepts, applied instead to magnetoassociation, that led to the observation [13] of collective Ramsey fringes between an atomic condensate and a small fraction of molecular condensate dressed by dissociated atom pairs [26–28].

The statistics of neutral atoms is determined by the number of neutrons in the nucleus, which must be odd for fermionic atoms and even for bosonic atoms. The sum of the total number of neutrons in the nucleus of the constituent atoms similarly determines the statistics of neutral molecules. Accordingly, molecules formed by free-bound association of two fermions will necessarily result in a boson, whereas fermionic molecules are born from the union of a boson and a fermion. Given a degenerate mixture of Bose and Fermi gases [14–16], is it possible that collective free-bound association could serve as a source of degenerate Fermi molecules?
To address this question, we model a degenerate Bose-Fermi mixture of atoms [14–16] coupled by either a Feshbach or photoassociation resonance to a Fermi-degenerate gas of molecules. The initial bosonic [fermionic] atoms are denoted by the field \( \phi(r,t) \) \( \psi_-(r,t), \) and the fermionic molecules by the field \( \psi_+(r,t) \). Neglecting particle-particle interactions, the Hamiltonian density for such an untrapped system can be written \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I, \) where

\[
\begin{align*}
\mathcal{H}_0 &= \frac{\hbar^2}{2m_B} \psi_+^+ \left[ -\frac{\hbar^2}{2m_-} - \mu_- \right] \psi_- \\
+ &\psi_+^+ \left[ -\frac{\hbar^2}{2m_+} + \delta_0 + \mu_+ \right] \psi_+, \\
\mathcal{H}_I &= -\frac{1}{2} \kappa \left( \psi_+ \psi_- \phi + \phi^\dagger \psi_+^\dagger \psi_- \right).
\end{align*}
\]

Here \( m_B \) \( (m_-) \) is the mass of a bosonic (fermionic) atom, \( m_+ = m_B + m_- \) is the mass of a molecule, \( \mu_{B(\pm)} \) is the so-called chemical potential that implicitly accounts for particle trapping, \( \delta_0 \) is the binding energy (detuning) of the molecular state relative to the dissociation threshold, and \( \kappa \) is the (real) atom-molecule coupling.

We now make some simplifications to allow for ease of modeling. First, we scale the Fermi (Bose) atom fields as \( \psi_+ \rightarrow e^{-i\mu_+ t} \psi_+ \) \( (\phi \rightarrow e^{-i\mu_B t} \phi) \), and the molecule field as \( \psi_+ \rightarrow e^{-i(\mu_+ + \mu_B) t} \psi_+ \), which yields \( \mu_+ \rightarrow \mu_+^* = \mu_+ - (\mu_+ + \mu_B) \); in turn, \( \mu_+^* \) is absorbed into the detuning. Second is that atom-molecule conversion occurs on a timescale much faster than the motion of the atoms in the trap, allowing us to neglect the kinetic energies, and justifying our omission of an explicit trapping potential for the particles. Third is that, whatever the Fermi energy associated with the fermionic components, it lies within the Wigner threshold regime, so that the coupling \( \kappa \) can be taken as the same for all modes. Finally, we switch to momentum space, but retain only the \( k = 0 \) atomic condensate modes since, due to Bose stimulation, these transitions are favored over \( k \neq 0 \) modes. The simplified Hamiltonian reads

\[
H = \sum_k \left[ \delta c_k^\dagger c_k - \frac{\kappa}{2} (c_k^\dagger b_k a + a^\dagger b_k^\dagger c_k) \right],
\]

where \( a \) annihilates a bosonic atom with wavevector \( k = 0 \) \( b_k \) \( (c_k) \) annihilates an atom (molecule) with wavevector \( k \), and \( \kappa = \kappa / \sqrt{N} \).

The system described by the Hamiltonian (2) is a particular nonlinear version of a two-level atom driven by an external field. Of course, the two-level atom is well known to undergo Rabi flopping [25], and similar behavior observed for the present system would in principle answer question of whether collective association will provide a shortcut to the formation of a Fermi-degenerate gas of molecules. We therefore consider an on-resonance system, for which \( \delta = 0 \) and the Heisenberg equations of motion are

\[
\begin{align*}
 i \dot{a} &= \kappa \sum_k b_k^\dagger c_k, \\
 i \dot{b}_k &= \kappa a^\dagger c_k, \\
 i \dot{c}_k &= \kappa b_k a.
\end{align*}
\]

If the system starts out with \( N_B \) bosonic atoms, the operator \( a \) will have a characteristic size \( \sqrt{N_B} \). On the other hand, while the fermionic operators \( b_k \) and \( c_k \) are characteristically of order unity, the sum over momentum states will ultimately introduce the initial number of fermionic atoms. Hence, for equal numbers of bosons and fermions, \( N_B = N_F = N \), it is intuitively obvious that the equations of motion (3) yield a system that evolves with the characteristic frequency \( \Omega = \sqrt{N} \kappa \).

Using Fock states, said intuition is confirmed by solving the time-dependent Schrödinger equation numerically, the results of which are shown in Fig. 1. For the simplest case of \( N = 1 \), the system undergoes complete conversion from a doubly-degenerate Fermi-Bose system of atoms to a Fermi-degenerate gas of molecules in a time \( \Omega^{-1} \). However, considering a larger number of initial atoms, \( N = 5 \), we see that quantum many-body fluctuations not only frustrate complete conversion, but also adjust the oscillation frequency and lead to collapse and revival. Increasing the initial particle number to \( N = 10 \) brings the amplitude of the first half-oscillation closer to the ideal case. This behavior is exactly analogous to the single-component bosonic case [19,21]. Although limited computational resources preclude the explicit investigation of large particle number, based on the bosonic analogy [19,21] we fully expect the first half oscillation to be complete for large \( N \), i.e., \( N \gg 1 \) initial Fermi atoms should be converted to \( N \) Fermi molecules over a timescale \( \tau \sim \Omega^{-1} \).

Nevertheless, collisions between particles will shift the energy of a state with a given number of atoms and molecules in a manner that depends nonlinearly on the numbers, making it difficult to maintain exact resonance. Furthermore, once a the system has Rabi flopped to molecules, the depopulated atomic condensate makes Bose stimulation, and thus neglect of the \( k \neq 0 \) BEC modes, questionable. And so we investigate the more robust possibility of rapid adiabatic passage [25]. From the Hamiltonian (2), it should be clear that the system will favor all atoms for large positive detunings, while favoring all molecules for large negative detunings. With \( \Omega \) established as the characteristic frequency for collective atom-molecule conversion, changes in the detuning that are of the order of \( \Omega \), and occur over a time of order \( \Omega^{-1} \), should qualify as adiabatic. Hence, if the detuning is swept in a linear fashion according to \( \delta = -\xi \Omega^2 t \), then dimensionless sweep rates \( \xi \ll 1 \) should enable rapid adiabatic passage to a degenerate gas of Fermi molecules.

Our suspicions are again corroborated by a Fock-state-based numerical solution to the Schrödinger equation, shown in Fig. 2. While many-body effects appear to rather seriously affect the efficiency of a marginally adi-
where the value of a atom-molecule, or Fermi-Fermi atom-molecule collisions in mind Bose-Bose atom, Fermi-Bose atom, Fermi-Bose relaxation induced by collisions with unconverted atoms. Brationally very hot, and therefore undergo vibrational transitions terms, and arises because the molecules are vibrationally very hot, and therefore undergo vibrational relaxation induced by collisions with unconverted atoms.

Before closing, we estimate some explicit numbers. We eschew photoassociation because of the losses associated with the electronically-excited state [30], and focus on the atom-molecule coupling provided by the $^{87}$Rb-$^{40}$K Feshbach resonance located at $B_0 = 534$ G [31], which has a width $\Delta R = 4$ G and an associated zero-field Fermi-Bose-atom scattering length $a_{FB}^K = -17.8$ nm. Accordingly, the atom-molecule coupling is $\mathcal{K} \approx (4\pi |\alpha_{FB}|^2 \mu_{Bohr} \Delta R m_\rho n_\rho)^{1/2} = 0.14 \text{cm}^{-1} \text{s}^{-1}$, where we have estimated the difference between the Fermi-Bose atom pair and molecular magnetic moments to be equal to the Bohr magneton $\mu_{Bohr}$. Assuming $N_B = 10^5$ condensate atoms in a trap with respective radial and axial frequencies $\omega_K/2\pi = 215$ Hz and $\omega_R/2\pi = 16.3$ Hz [16], the density of bosons is $\rho_B = 8.1 \times 10^{13} \text{cm}^{-3}$. As for the fermions, we assume a modest number, say, $N_F = 10^3$, which has three consequences: (i) the atomic BEC will act as a reservoir, thus absorbing any heat created by holes in the Fermi sea [32]; (ii) barring an unfortunately large scattering length for Bose-atom and Fermi-molecule collisions, we can neglect the possibility of any Fermi-Bose collapse instabilities [16]; (iii) the size of the Fermi cloud ($R_F = 8.3 \mu m$) is smaller than the BEC ($R_B = 10 \mu m$), so that overlap is not an issue. Moreover, for $N_B \gg N_F$, the timescale for atom-molecule conversion is $\tau_{m\rightarrow a} \sim (\sqrt{N_B} \mathcal{K})^{-1} = 8.2 \times 10^{-7} \text{s}$. This timescale is safely below the fastest timescale for trapped-atom motion $\tau_t = (\omega_K/2\pi)^{-1} = 4.7 \times 10^{-3} \text{s}$, justifying our neglect of trap dynamics and the kinetic energy; physically put, this means that the Fermi energy is negligible compared to the atom-molecule coupling strength.

Lastly we discuss the neglected role of particle-particle interactions, i.e., collisions. These are described in terms of a single parameter, the s-wave scattering length $a$, where the value of $a$ differs depending on whether one has in mind Bose-Bose atom, Fermi-Bose atom, Fermi-Bose atom-molecule, or Fermi-Fermi atom-molecule collisions (s-wave Fermi-Fermi atom and molecule collisions are of course Pauli blocked). Whatever the species involved, the real part of $a$ describes elastic collisions between particles, and leads to so-called mean-field shifts. The imaginary part need only be considered for atom-molecule collisions terms, and arises because the molecules are vibrationally very hot, and therefore undergo vibrational relaxation induced by collisions with unconverted atoms. The collisional coupling strength is then $\lambda = 2\pi \hbar a/m^*$, where $m^*$ is the mass of the atom or the reduced mass of the atom-atom (atom-molecule) pair, and it is included in the theory as a density dependent detuning. The units of the collisional coupling are $[\lambda] = \text{cm}^3/\text{s}$, so that an appropriate density factor is needed to arrive at a collisional rate. Estimating the real part of $a$ from the $^{87}$Rb-$^{40}$K collisions [31], and the imaginary part from collisions [22,24], a typical density ($\rho \sim 10^{14}$) leads to elastic and inelastic collisions that occur on a timescale $\tau_c \sim (\rho \lambda)^{-1} \approx 1$ ms. For rapid adiabatic passage, the contribution from the collisional interaction is negligible compared to the detuning, except near-resonance; however, the system only spends about $\tau_{a2m} \sim 1 \mu$s in this region, which is short enough to expect collisions to be negligible. If, by chance, collisions do turn out to be a problem, the trap could be expanded to further reduce the rate of collisions with unconverted atoms.

In conclusion, we highlight that the term $\Omega = \sqrt{N \kappa}$ was previously referred to as the Bose-enhanced free-bound coupling, and the detuning sweep, for example, referred to as Bose-stimulated rapid adiabatic passage from atoms to molecules. However, this behavior is now clearly independent of statistics, so that Bose stimulation of free-bound association has nothing whatsoever to do with Bose statistics, but is instead a many-body cooperative effect that applies equally well to Fermi-degenerate systems. We expect this situation to arise whenever the system is addressed as a unit, i.e., when the atom-molecule coupling strength is larger than the Fermi energy. To four-wave mixing in a Fermi gas of atoms [33,34], we therefore add a further example of fermions mimicking an effect that was previously attributed to Bose-Einstein statistics. Such behavior should allow for a shortcut to a degenerate gas of Fermi molecules via a collective photoassociation or Feshbach resonance.

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FIG. 1. Rabi-like oscillations in the fraction of Fermi-Bose atoms converted to Fermi molecules. The initial number of bosonic and fermionic atoms are equal, i.e., $N_B = N_F = N$. (a) The oscillations are complete for an initial number of atoms $N = 1$, while for $N = 5$ many-body effects lead to frequency-shifted oscillations that are incomplete and collapse. Better short-time agreement with the $N = 1$ result is obtained for $N = 10$. (b) The oscillations eventually revive.

FIG. 2. Rapid adiabatic passage from a Fermi-Bose gas of atoms to Fermi molecules. The detuning is swept as $\delta(t) = -\xi \Omega^2 t$, and $N_B = N_F = N$. (a) For borderline adiabaticity, $\xi = 1$, increasing the number of initial atoms from $N = 1$ to $N = 5$ indicates that many-body effects reduce the efficiency. (b) For $N = 5$ and $\xi = 0.1$, near-unit conversion is still possible, despite many-body effects.