Collective Molecule Formation in a Degenerate Fermi Gas via a Feshbach Resonance

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We model collisionless collective conversion of a degenerate Fermi gas of atoms into bosonic molecules via a Feshbach resonance, treating the bosonic molecules as a classical field and seeding the pairing amplitudes with random phases. A dynamical instability of the Fermi sea against association with molecules drives the conversion. The model qualitatively reproduces several experimental observations [Regal *et al.,* Nature (London) **424**, 47 (2003)]. We predict that the initial temperature of the Fermi gas sets the limit for the efficiency of atom-molecule conversion.

The idea [1] that an adiabatic sweep across an atom-molecule resonance can transform an atomic condensate into a molecular condensate has recently been ported to experiments on degenerate Fermi gases. By sweeping a magnetic field across a Feshbach resonance, at least part of the atoms have been demonstrably converted into molecules [2,3]. Magnetoassociation of atoms into molecules via a Feshbach resonance is also the key to experiments in which formation of a molecular condensate out of a degenerate Fermi gas has been observed [4].

To date, most experiments on magnetoassociation of fermionic atoms into molecules have been done in the collision-dominated regime. Collisions induce thermal equilibrium, and statistical mechanics, or indeed thermodynamics, seems to be the appropriate theoretical framework [5] (see also Cubizolles *et al.* [3]). An obvious exception is the adiabatic-sweep experiments of Ref. [2] on fast enough time scales that particle collisions are not a major factor [6]. These experiments are the domain of the time-dependent Schrödinger equation. Time-dependent association of a Bose-Einstein condensate of atoms into a condensate of molecules has attracted much interest [1,7]. Treating the condensates as classical fields, as opposed to quantum fields, guides and simplifies the analysis of Bose systems. In contrast, even the zero-temperature Fermi sea of atoms presumably cannot be represented as a classical field, a "macroscopic wave function." This is the technical dilemma we set out to tackle.

Here we develop a collisionless model for magnetoassociation of a two-component Fermi gas into bosonic molecules, treating the boson field classically. In this setting atom-molecule conversion builds up from a dynamical instability. We report on comparisons with experiments [2], and make the testable prediction that temperature limits the conversion efficiency in an adiabatic sweep of the magnetic field across the Feshbach resonance.

Consider a free two-component Fermi gas (spin-up and spin-down) with the annihilation operators $c_{\mathbf{k}\uparrow}$ and $c_{\mathbf{k}\downarrow}$ for states with momentum $\hbar\mathbf{k}$, and the corresponding Bose gas of diatomic molecules with annihilation operators $b_{\mathbf{k}}$. Absent collisions, the Hamiltonian reads

$$\frac{H}{\hbar} = \sum_{\mathbf{k}}[\epsilon_{\mathbf{k}}(c_{\mathbf{k}\uparrow}^{\dagger}c_{\mathbf{k}\uparrow} + c_{\mathbf{k}\downarrow}^{\dagger}c_{\mathbf{k}\downarrow}) + (\delta + \tfrac{1}{2}\epsilon_{\mathbf{k}})b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}}]$$
$$+ \sum_{\mathbf{k}\mathbf{k}'}[\kappa_{\mathbf{k}\mathbf{k}'}c_{\mathbf{k}\uparrow}^{\dagger}c_{\mathbf{k}'\downarrow}^{\dagger}b_{\mathbf{k}+\mathbf{k}'} + \text{H.c.}]. \qquad (1)$$

Here $\hbar\epsilon_{\mathbf{k}} \equiv \hbar\epsilon_k = \hbar^2 k^2/2m$ is the kinetic energy for an atom with mass m, $\hbar\delta$ is the atom-molecule energy difference that is adjusted by varying the magnetic field, and $\kappa_{\mathbf{k}\mathbf{k}'}$ are matrix elements for combining two atoms into a molecule. For s-wave processes, $\kappa_{\mathbf{k}\mathbf{k}'}$ are functions of the relative kinetic energy of the atom pair. The Hamiltonian (1) conserves the invariant particle number $N = \sum_{\mathbf{k}}(c_{\mathbf{k}\uparrow}^{\dagger}c_{\mathbf{k}\uparrow} + c_{\mathbf{k}\downarrow}^{\dagger}c_{\mathbf{k}\downarrow} + 2b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}})$. Given the quantization volume V, the invariant density is $\rho = N/V$.

In the spirit of classical field theory, the main assumption of our model is that the boson operators in the Heisenberg equations of motion are declared to be complex numbers. To facilitate the numerics, we furthermore keep only the molecular mode with $\mathbf{k} = 0$. We also assume that initially the occupation numbers of the spin-up and spin-down fermions are the same, and that the sample is rotationally invariant. The expectation values of the relevant operators then depend only on the energy of the state $\mathbf{k}$, $\hbar\epsilon = \hbar^2 k^2/2m$. We scale the c-number molecular amplitude as $\beta \equiv \sqrt{2/N}\, b_0$, define the fermion occupation numbers $P(\epsilon) = \langle c_{\mathbf{k}\uparrow}^{\dagger}c_{\mathbf{k}\uparrow}\rangle = \langle c_{\mathbf{k}\downarrow}^{\dagger}c_{\mathbf{k}\downarrow}\rangle$, the pairing or "anomalous" amplitudes $C(\epsilon) = \langle c_{-\mathbf{k}\downarrow}c_{\mathbf{k}\uparrow}\rangle$, and find

$$i\dot{C}(\epsilon) = 2\epsilon C(\epsilon) + \frac{1}{\sqrt{2}}\Omega f(\epsilon)\beta[1 - 2P(\epsilon)], \qquad (2)$$

$$i\dot{P}(\epsilon) = \frac{1}{\sqrt{2}}\Omega f(\epsilon)[\beta C^*(\epsilon) - \beta^* C(\epsilon)], \qquad (3)$$

$$i\dot{\beta} = \delta\beta + \frac{3\Omega}{2\sqrt{2}\,\epsilon_F^{3/2}}\int d\epsilon\,\sqrt{\epsilon}\,f(\epsilon)C(\epsilon). \qquad (4)$$

Here $\hbar\epsilon_F = \hbar^2(3\pi^2\rho)^{2/3}/2m$ is the usual Fermi energy. The energy-dependent atom-molecule coupling is $\kappa(\epsilon) = \kappa(0)f(\epsilon)$ with $f(0) = 1$, and the Rabi-like frequency is $\Omega = \sqrt{N}\kappa(0)$. As per Javanainen and Mackie [1], $\kappa(0) \propto 1/\sqrt{V}$, so that $\Omega \propto \sqrt{\rho}$. The integral arises from the continuum limit of the sum over $\mathbf{k}$, and $\sqrt{\epsilon}$ is a phase space factor responsible for the Wigner threshold law for the dissociation rate of molecules into atoms.

The problem with Eqs. (2)–(4) is that $\beta(t) = C(\epsilon, t) \equiv 0$ is always a solution. However, this solution may be unstable. To illustrate, we carry out the linear stability analysis of Eqs. (2)–(4) around the trivial solution for given occupation numbers $P(\epsilon)$. In the usual single-pole approximation that becomes increasingly accurate in the limit $\Omega \to 0$, and for $\delta > 0$, the Fourier transform of the small deviation from $\beta = C(\epsilon) = 0$ has the pole

$$\omega_0 = \delta - i\frac{3\pi\Omega^2}{8\epsilon_F^{3/2}}\sqrt{\frac{\delta}{2}}\left|f\left(\frac{\delta}{2}\right)\right|^2\left[1 - 2P\left(\frac{\delta}{2}\right)\right]. \qquad (5)$$

If the fermion occupation number satisfies $P(\epsilon) > \frac{1}{2}$ for some energy $\hbar\epsilon$, for a suitable detuning δ the evolution frequency has a positive imaginary part. The implication is that a small perturbation from the stationary state $\beta = C(\epsilon) = 0$ grows exponentially.

If dissociation of an isolated molecule into two atoms is energetically possible, it will invariably happen because the state space is much larger for two atoms than for a molecule. On the other hand, a filled Fermi sea of atoms may block dissociation. The state space of allowed molecular states is then the one that is larger, and the atoms are prone to *spontaneous magnetoassociation* into molecules. This is the nature of the instability.

The Fermi sea is thermodynamically unstable against the formation of Cooper pairs [8], and resonance superfluids [9] inherit an analog of this trait of BCS superconductors. Nonetheless, suggestive as the similarity may be, the BCS instability is different from the present one. Thermodynamical instability occurs because pairing lowers the energy, and so coupling to a reservoir with a low enough temperature leads to pairing. The hallmark of dynamical instability is that a small perturbation grows exponentially in time, environment or no environment.

Quantum fluctuations could trigger spontaneous magnetoassociation, but they are absent in our model. While much is known about the modeling of quantum fluctuations classically in a boson system [10], no corresponding general methods exist for fermions. We resort to the following heuristic device. Instead of starting out a calculation with zero anomalous amplitudes, we initially

seed them with random numbers having zero average; specifically, nonzero numbers with a random phase, $\langle c_{-\mathbf{k}\downarrow}c_{\mathbf{k}\uparrow}\rangle = e^{i\phi_{\mathbf{k}}}\langle c_{\mathbf{k}\uparrow}^\dagger c_{\mathbf{k}\uparrow}\rangle^{1/2}\langle c_{\mathbf{k}\downarrow}^\dagger c_{\mathbf{k}\downarrow}\rangle^{1/2}$. We then integrate Eqs. (2)–(4), whereupon the initial instability and the subsequent dynamics run their courses. We do the calculations for many choices of the random phases $\phi_{\mathbf{k}}$, and average the results. This procedure correctly reproduces the initial evolution $\propto t^2$ of the expectation value of the number of molecules in the full quantum model. The approximation lies in using it at all times. As a technical detail, in our numerical calculations we discretize $P(\epsilon)$ and $C(\epsilon)$ at equidistant points of ϵ separated by $\Delta\epsilon$, and resort to the analogous process.

Next we estimate the parameters of the model. First, atom-molecule conversion is unlikely to be the result of a contact interaction. Given a nonzero range, there is a cutoff in momentum/energy for the matrix element $\kappa_{\mathbf{k}\mathbf{k}'}$. We crudely assume that the coupling between atoms and molecules follows the Wigner threshold law up to the point when it abruptly cuts off at some energy $\hbar M$, and correspondingly set $f(\epsilon) = \theta(M - \epsilon)$. Second, we write the dependence of the detuning on the magnetic field as $\delta = \Delta\mu(B - B_0')/\hbar$, where B_0' is a tentative position of the Feshbach resonance and $\Delta\mu$ stands for the thus far unknown difference of the magnetic moment between a molecule and two atoms. Third, we have an atom-molecule coupling strength with the dimension of frequency, Ξ, defined by $\Omega = \Xi^{1/4}\hbar^{3/4}\sqrt{\rho}/m^{3/4}$.

We ignore the Fermi statistics by setting $P(\epsilon) \equiv 0$ in Eqs. (2)–(4). This yields a linear description of the coupling between molecules and atom pairs. With $\Delta\mu > 0$ and for detunings less than a threshold value δ_0 the remaining set of equations has a stationary solution; the Fourier transforms $\beta(\omega)$ and $C(\epsilon, \omega)$ have a real pole at a frequency $\omega(\delta)$ such that $\omega(\delta_0) = 0$ and $\omega(\delta) < 0$ for $\delta < \delta_0$. What in the absence of the coupling $\propto \Omega$ were a "bare" molecule and a pair of atoms become a "dressed" molecule. We interpret $\hbar\omega[\delta(B)]$ as the energy of the bound state of the dressed molecule for the given magnetic field, and shift the value of the resonance field from B_0' to B_0 so that $\delta_0 = 0$. The "renormalized" B_0 should equal the position of the Feshbach resonance in the limit of a dilute gas.

Finally, we fit the unknown parameters M, $\Delta\mu$, and Ξ to best reproduce the experimental binding energy of the molecule reported in Fig. 5 of Ref. [2]. The fit minimizes the relative error between the calculated and the measured values. The parameters are $M = 2\pi \times 100$ kHz, $\Delta\mu = 0.19\mu_B$, and $\Xi = 2\pi \times 580$ MHz.

By setting $P(\epsilon) = 0$ we have ignored the many-body shift of the Feshbach resonance. We could include the shift by allowing $P(\epsilon) \neq 0$. For our fitted parameters, experimental densities, and untouched zero-temperature Fermi sea, the shift could exceed one Gauss toward the direction of high B. The preparation of the sample in the experiments [2] quoted in our Fig. 1 alters $P(\epsilon)$ and
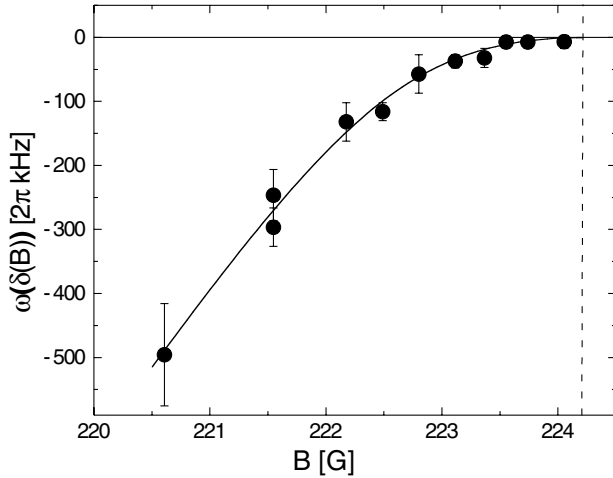
FIG. 1.   Energy of the bound state of the dressed molecule $\hbar\omega$ as a function of the magnetic field B from the experiments of Ref. [2] (filled circles) and from our calculations using the best-fit parameters M, Ξ, and $\Delta\mu$ (solid line).
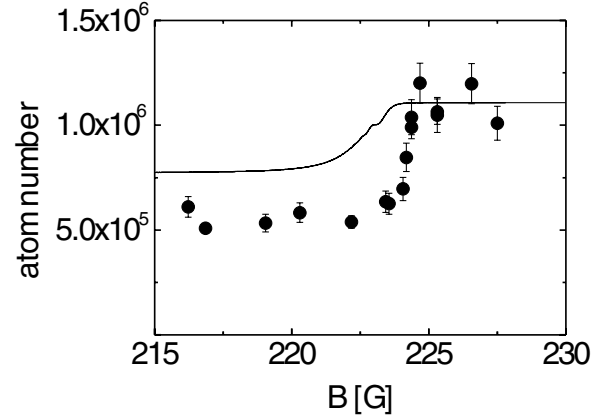

FIG. 2.   Experimental [2] (filled circles) and simulated (solid line) numbers of atoms remaining when the magnetic field is swept across the Feshbach resonance toward lower values. The simulations use the known experimental parameters and parameter values fitted as in Fig. 1.

modifies the shift in a manner that is difficult to account for self-consistently. Nonetheless, if we insert a full one-Gauss shift by hand and fit again, the most relevant parameter Ω increases by just 60%. Including or excluding the many-body shift of the Feshbach resonance should not make a qualitative difference in the values of the fitted parameters.

Armed with the parameter values, we next simulate a sweep of the magnetic field across the Feshbach resonance as in Fig. 1(a) of Ref. [2] by integrating Eqs. (2)–(4). From the experimental maximum density $\rho = 2.1 \times 10^{13}$ cm^{-3}, we have $\epsilon_F = 9.2 \times 2\pi$ kHz. The initial atomic occupation numbers $P(\epsilon)$ are set according to the experimental temperature, $k_B T/\hbar\epsilon_F = 0.21$. The discretization step for the atomic occupation numbers and anomalous amplitudes is $\Delta\epsilon = \epsilon_F/100$. We run the magnetic field sweep 64 times for different random phases of the anomalous amplitudes, and average the results. Figure 2 shows both the measured atom numbers (filled circles) and our calculations (solid line) as a function of the magnetic field when it is swept at the rate $(40 \ \mu\text{s/G})^{-1}$.

Experimentally, half of the atoms are converted into molecules at $(40 \ \mu\text{s/G})^{-1}$, while our calculations give a 30% conversion. There are a number of reasons why a full quantitative agreement cannot be expected, e.g., keeping only one molecular mode, using the maximum density of the atoms instead of making an allowance for the density distribution in the trap, simplistic modeling of the energy dependence of the atom-molecule coupling $f(\epsilon)$, and ignoring many-body shifts while fitting the parameters. Nonetheless, our model seems to identify the correct physics parameters, first and foremost the Rabi-like frequency Ω, and gives a reasonable estimate for their values.

In the experiments [2] the magnetic field was also swept back and forth, whereupon the molecules dissociate back into atoms. With the random initial phases of the anomalous amplitudes, it is not obvious that our simulations should reproduce this feat, but they do.

The puzzling feature of the experiments [2] is that, no matter how slow the sweep rate, the conversion efficiency is limited to about 50%. To investigate, we carry out magnetic field sweeps as in Fig. 2, except that the sweep rate is such that the detuning as a function of time behaves as $\delta = -\xi\Omega^2 t$ with $\xi = 0.05$, corresponding to $\dot{B} = (400 \ \mu\text{s/G})^{-1}$. If Ω^{-1} determines the time scale, for $\xi = 0.05$ one would expect adiabaticity. We vary the temperature and plot the final conversion efficiency as a function of the temperature. The results are shown in Fig. 3. At $T = 0$, 98% of the atoms are converted into molecules. However, by the time the temperature has reached the
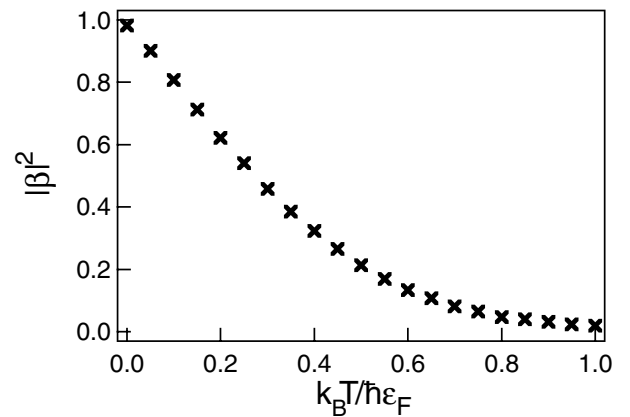

FIG. 3.   Fraction of atoms converted into molecules in a sweep such as in Fig. 2, except that here the sweep rate is slower, $(400 \ \mu\text{s/G})^{-1}$, and the results are plotted as a function of the varying initial temperature of the atomic gas.

Fermi temperature, $k_B T = \hbar \epsilon_F$, the conversion efficiency has dropped to 2%. At the typical experimental temperatures with $k_B T / \hbar \epsilon_F \sim 0.2$–$0.3$, the efficiency, in fact, hovers in the neighborhood of 50%.

For an increasing temperature, a decreasing number of the initial atomic states have a thermal occupation number of at least $\frac{1}{2}$. The instability responsible for initiating atom-molecule conversion then acts on fewer and fewer atomic modes. We presume this is the reason for the temperature dependence.

The weakest point in our argument may be the assumption of a single molecular mode, according to which the molecules emerge as a condensate. It would appear that, without any seed for the molecular condensate or initial phase from the atoms, the atoms are converted into a coherent molecular condensate in a time that is independent of the size of the sample. This can hardly be correct. Inclusion of a multitude of molecular modes would probably cure this shortcoming. By construction we would still have a classical field representing the molecules, but it could consist of patches with uncorrelated phases. A numerical modeling of this situation is in principle possible, but much more demanding than our present calculations.

Instead, we present an estimate for the size of the patches. The natural velocity parameter in this problem is the Fermi velocity, $v_F = \sqrt{2\hbar \epsilon_F / m}$. Suppose the conversion takes place in a time Δt, then atoms in a region of size $\ell \sim v_F \Delta t$ would plausibly be able to form a patch of molecular condensate. Let us estimate the conversion time from Fig. 2, say, as the difference between the times when the number of molecules rises from $\frac{1}{4}$ to $\frac{3}{4}$ of its final value in the dashed-line data, then the size of the patch and the characteristic distance between the atoms are related by $\ell \sim 2\rho^{-1/3}$. Our modeling should be adequate if it is taken to represent a patch of about $2^3 \sim 10$ atoms. Within each patch we could still resort to the single mode approximation and use the Bose-enhanced coupling strength Ω that depends on density but not directly on the number of atoms. However, the patchy molecular condensate would not appear as a condensate in an experiment, but would display a momentum distribution not narrower than $\sim \hbar/\ell$.

The number of atoms in a zero-temperature Fermi sea is the same as the number of occupied states, so for consistency we should use a step of the order of $\Delta\epsilon \sim \epsilon_F/10$ in our modeling. We have used $\Delta\epsilon = \epsilon_F/100$ in Figs. 2 and 3. Now, constants of the order of unity are beyond our dimensional-analysis argument, and the step size estimate should be taken with a grain of salt. On the other hand, numerically, the step size dependence is logarithmic and weak. Simulations with just one molecular mode should therefore be valid semiquantitatively.

Starting from a microscopic many-particle theory, we have modeled collisionless collective conversion of a degenerate Fermi gas into bosonic molecules via a Feshbach resonance. The key techniques are to treat the molecules as a classical field, and to seed pairing amplitudes with random phases. The main concept we have unearthed is dynamical instability of a Fermi sea against magnetoassociation into molecules. The same instability should occur in photoassociation. Our model reproduces qualitatively all experimental observations [2] where we have tried a comparison. Moreover, we have the testable prediction that temperature sets the limit for the efficiency of atom-molecule conversion.

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[1] E. Timmermans *et al.*, cond-mat/9805323; J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999); F. A. van Abeelen and B. J. Verhaar, Phys. Rev. Lett. **83**, 1550 (1999); F. H. Mies, E. Tiesinga, and P. S. Julienne, Phys. Rev. A **61**, 022721 (2000).

[2] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature (London) **424**, 47 (2003).

[3] K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003); J. Cubizolles *et al.*, Phys. Rev. Lett. **91**, 240401 (2003).

[4] S. Jochim *et al.*, Science **302**, 2101 (2003); M. Greiner, C. A. Regal, and D. S. Jin, Nature (London) **426**, 537 (2003); M. W. Zwierlein *et al.*, Phys. Rev. Lett. **91**, 250401 (2003).

[5] L. D. Carr, G. V. Shlyapnikov, and Y. Castin, Phys. Rev. Lett. **92**, 150404 (2004).

[6] Here, of course, we do *not* think of the process of atom-molecule conversion as a collision.

[7] S. J. J. M. F. Kokkelmans and M. J. Holland, Phys. Rev. Lett. **89**, 180401 (2002); M. Mackie, K.-A. Suominen, and J. Javanainen, Phys. Rev. Lett. **89**, 180403 (2002); T. Köhler, T. Gasenzer, and K. Burnett, Phys. Rev. A **67**, 013601 (2003); R. A. Duine and H. T. C. Stoof, J. Opt. B **5**, S212 (2003); K. V. Kheruntsyan and P. D. Drummond, Phys. Rev. A **66**, 031602 (2002); V. A. Yurovsky and A. Ben-Reuven, Phys. Rev. A **67**, 050701 (2003).

[8] L. N. Cooper, Phys. Rev. **104**, 1189 (1956).

[9] E. Timmermans, K. Furuya, P. W. Milonni, and A. K. Kerman, Phys. Lett. A **285**, 228 (2001); M. Holland, S. J. J. M. F. Kokkelmans, M. L. Chiofalo, and R. Walser, Phys. Rev. Lett. **87**, 120406 (2001); Y. Ohashi and A. Griffin, Phys. Rev. Lett. **89**, 130402 (2002).

[10] C. W. Gardiner and P. Zoller, *Quantum Noise* (Springer, Berlin, 2000), 2nd ed.