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New directions in degenerate dipolar molecules via collective association

M. Mackie[1,a], O. Dannenberg[2], J. Piilo[3], K.-A. Suominen[2,4], and J. Javanainen[5]

[1] QUANTOP–Danish National Research Foundation Center for Quantum Optics, Department of Physics and Astronomy, University of Aarhus, 8000 Aarhus C, Denmark
[2] Helsinki Institute of Physics, PL 64, 00014 Helsingin yliopisto, Finland
[3] School of Pure and Applied Physics, University of KwaZulu-Natal, Durban 4041, South Africa
[4] Department of Physics, University of Turku, 20014 Turun yliopisto, Finland
[5] Department of Physics, University of Connecticut, Storrs, Connecticut, 06269-3046, USA

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Abstract. We survey results on the creation of heteronuclear Fermi molecules by tuning a degenerate Bose-Fermi mixture into the neighborhood of an association resonance, either photoassociation or Feshbach, as well as the subsequent prospects for Cooper-like pairing between atoms and molecules. In the simplest case of only one molecular state, corresponding to either a Feshbach resonance or one-color photoassociation, the system displays Rabi oscillations and rapid adiabatic passage between a Bose-Fermi mixture of atoms and fermionic molecules. For two-color photoassociation, the system admits stimulated Raman adiabatic passage (STIRAP) from a Bose-Fermi mixture of atoms to stable Fermi molecules, even in the presence of particle-particle interactions. By tailoring the STIRAP sequence it is possible to deliberately convert only a fraction of the initial atoms, leaving a finite fraction of bosons behind to induce atom-molecule Cooper pairing via density fluctuations; unfortunately, this enhancement is insufficient to achieve a superfluid transition with present ultracold technology. We therefore propose the use of an association resonance that converts atoms and diatomic molecules (dimers) into triatomic molecules (trimers), which leads to a crossover from a Bose-Einstein condensate of trimers to atom-dimer Cooper pairs. Because heteronuclear dimers may possess a permanent electric dipole moment, this overall system presents an opportunity to investigate novel microscopic physics.

PACS. 03.75.Ss Degenerate Fermi gases – 05.30.Fk Fermion systems and electron gas – 34.10.+x General theories and models of atomic and molecular collisions and interactions (including statistical theories, transition state, stochastic and trajectory models, etc.) – 74.20.Mn Nonconventional mechanisms (spin fluctuations, polarons and bipolarons, resonating valence bond model, anyon mechanism, marginal Fermi liquid, Luttinger liquid, etc.) – 21.10.-k Properties of nuclei; nuclear energy levels

1 Introduction

It has recently come to the fore that collective association can actually produce a quantum degenerate molecules, serving as a compliment to the development of buffer-gas cooling [1] and Stark-deceleration [2] techniques. The trail to molecular Bose-Einstein condensate (BEC) was pioneered by experiments [3] with photoassociation of ^{87}Rb condensate that were just on the verge [4] of coherent atom-molecule conversion. Next-generation experiments were aimed at the strongly interacting regime [5,6], and probed the predicted [4,7,8] photoassociation rate limit. As of late, laser experiments [9] have confirmed [4,10,11] an optically tunable BEC scattering length, no small feat in the face of rapid spontaneous decay. Meanwhile, groundbreaking magnetoassociation experiments demonstrated a magnetically tunable scattering length [12,13], and led to the controlled collapse of a condensate with atom bursts emanating from remnant condensate [14]. Subsequent coherent oscillations between burst and remnant [15] were interpreted as evidence for atom-molecule coherence [16–19]. In the search for the neutral-atom analogue of superconducting Cooper pairs of electrons [20,21], ultracold [22–24] and condensate [25–27] molecules were unambiguously created via magnetoassociation of fermionic atoms [28–30]. Similarly, quantum degenerate molecules have been observed and characterized in magnetoassociation of atomic BEC [31].

Collective association is a general theory to describe a quantum-degenerate gas tuned near a photoassociation or Feshbach resonance. The former is a resonance where two colliding atoms absorb a photon and jump to a bound molecular state [32], whereas the latter is a resonance where one atom from a colliding pair spin flips in the

[a] e-mail: mattmackie2000@yahoo.com

presence of a magnetic field [33,34], known affectionately as magnetoassociation. Some of the first theoretical investigations into association of a quantum degenerate gas pointed to strongly enhanced molecule formation [35,36], and a focus on Bose-Einstein condensates [37–43] in particular led to the idea of Bose-enhanced molecule production [44], or so-called superchemistry [45]. However, it soon became clear that this idea applied equally well to degenerate Fermi and Bose-Fermi mixtures, indicating the possibility for changes in statistics, i.e., creating Bose molecules from Fermi mixtures [46–49,52] or Fermi molecules from Bose-Fermi mixtures [50,51], and thereby taking superchemistry to a new level. Moreover, collective association [54] of Bose-Fermi mixtures [55] in particular offers a convenient source of ultracold polar molecules, which can be used for quantum computation [57] and searches for the standard-model-violating electric dipole moment of the electron [58].

So motivated, our attention has therefore turned to resonant association in Bose-Fermi mixtures of atoms, and the subsequent possibility of atom-molecule Cooper pairing. While the Raman formation of ground state Fermi molecules appears feasible in practice [52], the subsequent off-resonant transition to a superfluid of atom-molecule Cooper pairs occurs out of reach of present ultracold technology [52]; hence, an association resonance for *trimers* is apparently necessary to experimentally realize atom-dimer Cooper pairing [53]. The purpose of this article is to survey our results on collective Fermi molecule formation, in order to highlight future research avenues in the physics of quantum degenerate dipolar gases.

The survey herein is outlined as follows. In Section 2, we discuss the broad features of converting a degenerate Bose-Fermi mixture into Fermi molecules via a collective association resonance, in particular Rabi-like oscillation and rapid adiabatic passage. In Section 3, the focus is on the formation of stable Fermi molecules with Raman photoassociation of a Bose-Fermi mixture, and the subsequent possibility for boson-induced Cooper pairing between atoms and molecules, where Raman photoassociation converts a fraction of the initial mixture into molecules and the leftover bosons enhance the atom-molecule interaction. Since this enhancement is insufficient to induce pairing within reach of present ultracold technology, Section 4 explores the possibility of driving atom-molecule Cooper pairing with an association resonance that converts atom-dimer pairs into trimers, i.e., a crossover from a BEC of trimers to atom-dimer Cooper pairs. Besides a summary, Section 5 identifies some open questions regarding dipolar particle interactions.

2 Shortcut to degenerate Fermi molecules via collective association

Analogous with coherent optical transients in few level atomic systems [59], photoassociation of a BEC has been predicted to induce Rabi-like oscillations between atomic and molecular condensates [38,39,45], 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 above threshold) and all molecules for large negative detunings (far below 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 [38]. Incidentally, it was a particular combination of these two concepts, applied instead to magnetoassociation, that led to the observation [15] of collective Ramsey fringes between an atomic condensate and a small fraction of molecular condensate dressed by dissociated atom pairs [16–19]. Given a degenerate mixture of Bose and Fermi gases [55], will similar coherent phenomena manifest in free-bound association degenerate Fermi molecules?

To address this question, we model an ideal degenerate Bose-Fermi mixture of atoms coupled by either a Feshbach or photoassociation resonance to a Fermi-degenerate gas of molecules. But first we make some simplifications to allow for ease of modeling. First, by making a proper [50] unitary transformation, any explicit chemical potential can be absorbed into the detuning and forgotten. Second, collective association occurs on a timescale much faster than trapped-atom motion, allowing neglect of the kinetic energies, and justifying omission of an explicit trap. Third, any Fermi energies lie within the Wigner threshold regime, so that the coupling $\mathcal{K}$ can be taken as the same for all modes. Finally, we work in momentum space, but retain only the $\mathbf{k} = 0$ condensate modes since Bose stimulation favors these transitions over $\mathbf{k} \neq 0$ modes. The simplified Hamiltonian reads

$$H = \sum_{\mathbf{k}} \left[\delta b_{\mathbf{k}}^\dagger b_{\mathbf{k}} - \tfrac{1}{2}\kappa (b_{\mathbf{k}}^\dagger a_{\mathbf{k}} c + c^\dagger a_{\mathbf{k}}^\dagger b_{\mathbf{k}}) \right], \qquad (1)$$

where c annihilates a condensate atom, $a_{\mathbf{k}}$ ($b_{\mathbf{k}}$) annihilates an atom (molecule) with wavevector $\mathbf{k}$, and $\kappa = \mathcal{K}/\sqrt{V}$.

From the Heisenberg equations of motion for an on-resonance system ($\delta = 0$) [50], it is easy to show that for equal numbers of bosonic and fermionic atoms, $N_B = N_F = N$, the system evolves with the characteristic frequency $\Omega = \sqrt{N}\kappa$. Using Fock states, said intuition is confirmed by solving the Schrödinger equation numerically, the results of which are shown in Figure 1. For the two-body case ($N = 1$), a complete oscillation between Fermi-Bose atoms and Fermi molecules occurs in a time $2\pi/\Omega$. However, for $N = 5$ quantum many-body fluctuations not only frustrate complete conversion, but also shift the oscillation frequency and lead to collapse and revival. Increasing the initial particle number to $N = 10$ gives better short-time agreement with the two-body result. This behavior is exactly analogous to the single-component bosonic case [38,39]. Although limited computational resources preclude explicit investigation, based on the bosonic analogy [38,39] we fully expect the first half oscillation to be complete for large particle number.

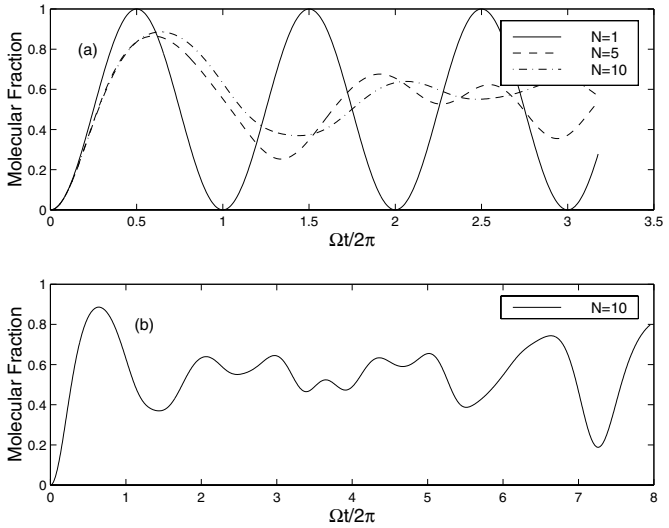

Fig. 1. Rabi-like oscillations in the fraction of a Fermi-Bose gas of atoms converted to Fermi molecules. The initial number of bosonic and fermionic atoms are equal, i.e., $N_B = N_F = N$. (a) The oscillations are complete for an initial number of atoms $N = 1$, while for $N = 5$ many-body effects lead 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.

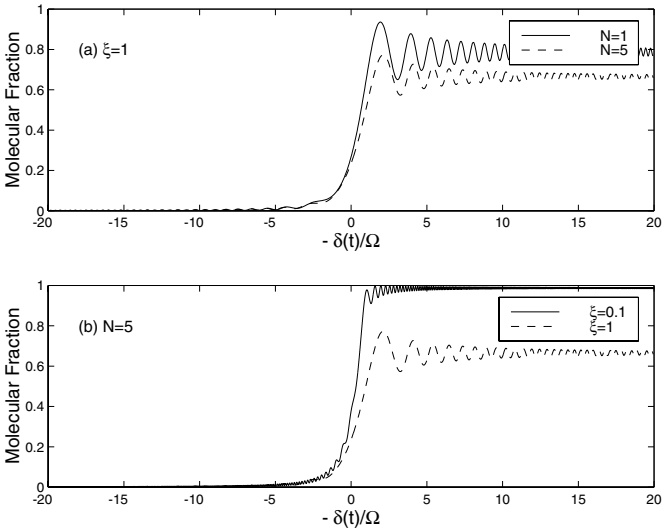

Fig. 2. Rapid adiabatic passage from a Fermi-Bose gas of atoms to Fermi molecules. The detuning is swept as $\delta(t) = -\xi\Omega^2 t$, and $N_B = N_F = N$. (a) For borderline adiabaticity, $\xi = 1$, increasing the number of initial atoms from $N = 1$ to $N = 5$ indicates that many-body effects reduce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

From the Hamiltonian (1), it should also be clear that the system will favor all atoms for large positive detunings, while favoring all molecules for large negative detunings. With Ω established as the characteristic frequency for collective atom-molecule conversion, changes in the detuning that are of the order of Ω, and occur over a time of order Ω^{-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 $\xi \lesssim 1$ should enable rapid adiabatic passage to degenerate Fermi molecules. Our suspicions are again corroborated by a Fock-state-based numerical solution to the Schrödinger equation, shown in Figure 2. While many-body effects appear to rather seriously affect the efficiency of a marginally adiabatic sweep ($\xi = 1$) compared to the $N = 1$ case, the difference between $N = 5$ and $N = 10$ (not shown) is in fact small. Overall, many-body effects are expected to be weak for near-adiabatic sweeps ($\xi \sim 1$), and vanishing for sweeps that are deep-adiabatic ($\xi \lesssim 0.1$), again in agreement with our BEC experience [38].

For an estimate of explicit numbers we eschew photoassociation because of the losses associated with the electronically-excited state, and focus on the atom-molecule coupling provided by the Feshbach resonance located at $B_0 = 534$ G [61], which has a width $\Delta_R = 4$ G and an associated zero-field Fermi-Bose-atom scattering length $a_{aa}^{FB} = -17.8$ nm. Accordingly, the atom-molecule coupling is $\mathcal{K} \approx (4\pi|a_{aa}^{FB}|\mu_{\rm Bohr}\Delta_R/m_r)^{1/2} = 0.14$ cm$^{3/2}$ 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 $\mu_{\rm Bohr}$. Assuming $N_B = 10^5$ condensate atoms in a trap with respective radial and axial frequencies $\omega_r/2\pi = 215$ Hz and $\omega_a/2\pi = 16.3$ Hz [56], the density of bosons is $\rho_B = 8.1 \times 10^{13}$ 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 [62]; (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 [56]; (iii) the size of the Fermi cloud ($R_F = 8.3$ μm) is smaller than the BEC ($R_B = 10$ μm), so that overlap is not an issue. Moreover, for $N_B \gg N_F$, the timescale for atom-molecule conversion is $\tau_{a2m} \sim (\sqrt{\rho_B}\,\mathcal{K})^{-1} = 8.2 \times 10^{-7}$ s. This timescale is safely below the fastest timescale for trapped-atom motion $\tau_t = (\omega_r/2\pi)^{-1} = 4.7 \times 10^{-3}$ 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.

It is important to mention two neglected complications: noncondensate modes and interactions between particles. On the matter of dissociation to noncondensate modes, and the related pair correlations [16–19], we note that in the all-boson case [60] such transitions can be neglected for a sweep directed as in Figure 2, i.e., for $\dot{\delta} < 0$. Given the success of the analogy so far, similar is expected for the Fermi molecule system. As for collisions between particles, they are described in terms of the coupling strength is $\lambda = 2\pi\hbar a/m^*$, where a is the s-wave scattering length, m^* is the mass of the atom or the reduced mass of the atom-atom (atom-molecule) pair. An estimate for λ indicates that it is negligible compared to

the atom-molecule coupling for a typical ^{87}Rb–^{40}K system and, besides, particle interactions have little effect on sweeps across resonance in the boson case [63] and similar is expected for Fermi molecules.

3 Raman photoassociation of stable Fermi molecules and their subsequent Cooper pairing with atoms

Given that a degenerate mixture of Bose and Fermi atoms admits basic collective association, the natural question to ask is whether Raman photoassociation to the ground molecular state will be efficient, since for bosons it is not a trivial matter to overcome the condensate mean-field shifts. If so, the follow up to this question is whether it possible to realize Cooper pairing, not between atoms or molecules, but instead between atoms and molecules.

First introduced to explain superconductivity, anomalous quantum correlations between two degenerate electrons with equal and opposite momenta — Cooper pairs — are due physically to an electron-electron attraction mediated by the exchange of lattice-vibration-generated phonons [64], and are a manifestation of fermionic superfluidity [65]. Anomalous pairing between different chemical species was immediately suggested to explain the larger excitation energy for nuclei with even rather than odd numbers of nucleons [66], although it turned out that interspecies pairing plays the dominant role. Today quantum matter optics offers a means to explore condensed-matter and nuclear physics by proxy, such as the pairing of fermions in atomic traps and nuclei [67]. Here we point out that, while the dipolar-dipolar case has been considered, Cooper pairing between non-dipolar atoms and dipolar molecules offers a novel avenue of investigation.

In this section we survey Raman photoassociation [44,68–70] of Bose-Fermi mixtures of atoms [55], and the subsequent prospects for Cooper pairing between different chemical species (i.e., atoms and molecules). First, we demonstrate that an arbitrary number of stable Fermi molecules can be created via fractional [71] stimulated Raman adiabatic passage (STIRAP [72]), which is feasible because, contrary to bosonic systems [69], collisional interactions can be negligible. Density fluctuations in the condensate leftover from the photoassociation process then replace the vibrating ion lattice of the superconductor [73], and the subsequent phonon exchange can enhance the intrafermion attraction. We find that a typical attraction is enhanced, but this enhancement is insufficient for a transition to atom-molecule Cooper pairs within reach of present ultracold technology.

We model a Bose-Fermi mixture of atoms coupled by heteronuclear photoassociation to electronically-excited Fermi molecules, which is favored over homonuclear transitions for well resolved resonances. The excited molecules are themselves coupled by a second laser to electronically stable molecules. For a degenerate system, the bosonic [fermionic] atoms of mass m_0 [m_+] are represented by the field $\psi_0(\mathbf{r},t)$ [$\psi_+(\mathbf{r},t)$], and the excited [stable] fermionic molecules of mass $m_e = m_0 + m_+$ [$m_- = m_e$] by the field $\psi_e(\mathbf{r},t)$ [$\psi_-(\mathbf{r},t)$], with the boson (fermion) field operator obeying commutation (anticommutation) relations. The Hamiltonian density for said non-ideal system is $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I$, where

$$\frac{\mathcal{H}_0}{\hbar} = -\Delta\psi_-^\dagger\psi_- + (\delta-\Delta)\psi_e^\dagger\psi_e + \lambda_{+-}\psi_+^\dagger\psi_-^\dagger\psi_-\psi_+ + \sum_\sigma \psi_\sigma^\dagger\left[-\frac{\hbar\nabla^2}{m_\sigma} - \mu_\sigma + \lambda_{0\sigma}\psi_0^\dagger\psi_0\right]\psi_\sigma\,, \tag{2a}$$

$$\frac{\mathcal{H}_I}{\hbar} = -\tfrac{1}{2}\left[\left(\mathcal{K}_+\psi_e^\dagger\psi_+\psi_0 + \Omega_-\psi_e\psi_-^\dagger\right) + \text{H.c.}\right]. \tag{2b}$$

The light-matter coupling due to laser 1 (2) is $\mathcal{K}_+$ (Ω_-), and the intermediate (two-photon) laser detuning, basically the binding energy of the excited (stable) molecular state relative to the photodissociation threshold, is δ (Δ). Particle trapping is implicit to the chemical potential $\hbar\mu_\sigma$ ($\sigma = 0, e, \pm$), and explicit traps can be neglected for most practical purposes. Low-energy (s-wave) collisions are accounted for by the boson-boson (boson-fermion, fermion-fermion) interaction strength $\lambda_{00} = 2\pi\hbar a_{00}/m_0$ ($\lambda_{0\pm} = 4\pi\hbar a_{0\pm}/m_{0\pm}^*$, $\lambda_{+-} = 4\pi\hbar a_{+-}/m_{+-}^*$), with $a_{\sigma_1\sigma_2}$ the s-wave scattering length and $m_{\sigma_1\sigma_2}^*$ the reduced mass. Spontaneous decay, included as $\Im[\delta] = -\Gamma$, is generally large enough to justify the exclusion of excited-molecule collisions.

The backbone of free-bound-bound stimulated Raman adiabatic passage is *counterintuitive* pulse timing: the two lasers are adjusted so that the level with the most population is less strongly coupled to the electronically-excited intermediate state, i.e., the bound-bound pulse arrives first and the free-bound pulse arrives last, so that the intermediate state population and the associated losses are kept to a minimum. When the number of bosons is much greater than the number of fermions, $N_B \gg N_F$, the frequency scale for atom-molecule conversion is set by $\Omega_+ = \sqrt{\rho_B}\mathcal{K}_+$. Although a maximum $N_B = 100$ is used, qualitative scaling to large boson number is cinched by assuming a density, $\rho_B = 5\times10^{14}$ cm^{-3}, consistent with $N_B = 1.3\times10^6$ Bose-condensed ^{85}Rb atoms in a trap with radial and axial frequencies $\omega_r = 100\times2\pi$ Hz and $\omega_a = 10\times2\pi$ Hz. For this density, a ballpark peak value for the atom-molecule coupling is $\Omega_+ \sim \Omega_0 = 2\pi$ MHz. A typical spontaneous decay rate is $\Gamma = 10\times2\pi$ MHz. The (assumedly negative) Fermi atom-molecule scattering length is estimated as $|a_{+-}| = a_{00} = 5.29$ nm. The number of fermions is restricted to $N_F = 4$ for numerical ease, and large-particle-number scaling is again ensured by picking a density, $\rho_F = 1.1\times10^{12}$ cm^{-3}, consistent with $N_F = 5\times10^3$ ^{40}K atoms in the same (mass-adjusted) trap as the bosons, so that $\Lambda_{+-} = \lambda_{+-}/V = 5.81\times2\pi$ Hz. Numerics are further eased by restricting collisions between fermions to a bare minimum. Also, $N_B \gg N_F$ means that collisions with condensate atoms can be neglected. See reference [50] for further details.

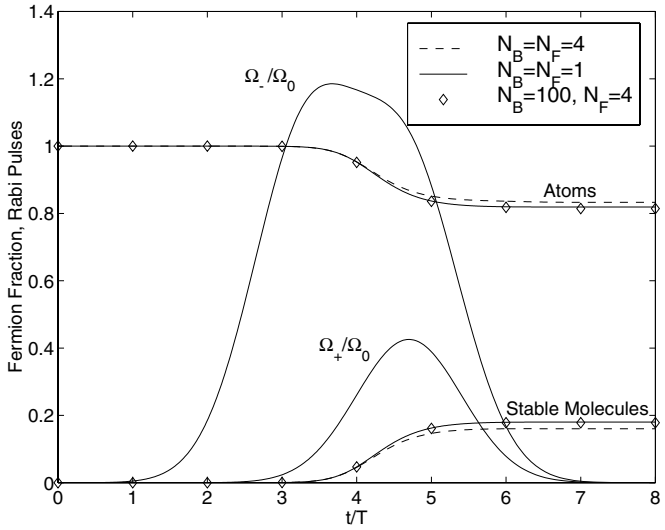

Fig. 3. Creation of an arbitrary mixture of Fermi degenerate atoms and molecules via fractional stimulated Raman adiabatic passage. Units of frequency are set by the choice $\Omega_0 = 2\pi$ MHz $= 1$, and the pulse parameters are $\alpha = 0.14\pi$, $T = 5 \times 10^3$, and $\tau = 0.7T$. For $N_B = 100$ and $N_F = 4$, fractional STIRAP exhibits no visible difference from the $N_B = N_F = 1$ case, while $N_B = N_F = 4$ illustrates that many-body effects can limit the conversion efficiency.

Of course, complete conversion would leave no atoms to form Cooper pairs with molecules, so we pursue fractional STIRAP [71] via the Vitanov et al. [71] pulseforms: $\Omega_+(t) = \Omega_0 \sin\alpha \exp[-(t-\tau)^2/T^2]$ and $\Omega_-(t) = \Omega_0 \exp[-(t+\tau)^2/T^2] \quad + \quad \Omega_0 \cos\alpha \exp[-(t-\tau)^2/T^2]$, where $\tan\alpha$ sets the final population fraction. Using Fock states and a Hamiltonian derived from equation (2b) [52], we solve the Schrödinger equation numerically. The key results are presented in Figure 3. For $N_F = 4$ and $N_B = 100$, the system reproduces the single particle case ($N_B = N_F = 1$), i.e., the results are identical to those for a three-level atom, as expected for a mostly-undepleted boson field. Atom-molecule collisions are negligible for couplings as large as $\Lambda_{+-}/\Omega_0 = 10^{-5}$, which is similar to a bosonic system, lending confidence to the restricted-collision model. Furthermore, $N_F = N_B = 4$ shows that many-body effects can limit molecular conversion. This many-body effect is similar to the one-color BEC case [38, 39], and is not attributable to fermion statistics.

The important point is that STIRAP works basically as expected, *even in the presence of collisions*. This situation arises because, when $N_B \gg N_F$, the condensate density is effectively fixed, and the associated mean-field contribution simply amounts to a static bias that can be absorbed into the detuning; with Fermi-Fermi collisions blocked, which is most likely for small ground-state molecules, only collisions between the Fermi atoms and molecules can move the system off the required two-photon resonance, and STIRAP works better. In other words, we get the advantage of Bose enhancement of the free-bound coupling ($\Omega_+ \propto \sqrt{\rho_B}$), without the mean-field shifts. While limited computational resources preclude explicit investigation, these results should scale qualitatively with increasing particle number, as for up to $N_F = 20$ in one-color production of Fermi molecules.

Now we are safe to presume the existence of an arbitrary admixture of Fermi-degenerate atoms and molecules, and thus to consider any subsequent anomalous pairing. Once the transient STIRAP pulses have vanished, the system is described by $\mathcal{H}_0$ [Eq. (2a)] with $\Delta = \delta = 0$ and $\sigma = 0, \pm$. For equal-mass fermions, it is known that a fermion density fluctuation gives rise to an effective chemical potential for the bosons, which creates a bosonic density fluctuation, which in turn leads to an effective chemical potential for the fermions. In other words, phonons spawned by BEC density fluctuations are exchanged between the fermions, altering their interaction. Just like lattice vibrations that drive the attraction between degenerate electrons in superconductors, BEC density fluctuations lead to an attractive interaction that can enhance overall attractions, and thus Cooper pair formation [73].

Here the effective Fermi-Fermi scattering length is

$$\bar{a}_{+-} = a_{+-}\left[1 + \frac{\ln(4e)^{2/3}}{\pi} k_F a_{+-} - H\,\frac{\lambda_{0+}\lambda_{0-}}{\lambda_{00}\lambda_{+-}}\right], \quad (3)$$

where $H = \ln(1+x^2)/x^2$ with $x = \hbar k_F/m_0 v_s$ and $v_s = (\rho_B \hbar \lambda_{00}/m_0)^{1/2}$ is the speed of phonons in BEC; $\hbar k_F \ll m_0 v_s$ implies $H \approx 1$. In other words, the effective scattering length can be written $\bar{a}_{+-}/a_{+-} = 1 + \eta_{FF} - \eta_{FB}$, where η_{FF} (η_{FB}) is the contribution to atom-molecule interactions from fermion-fermion (boson-fermion) fluctuations. Implicit to expression (3) is the perturbative assumption $\eta_{FB} \ll \eta_{FF}$. The immediate contrast with reference [73] is that $\eta_{FB} < 0$ is allowed. For a weakly attractive system ($k_F|a_{\sigma_1\sigma_2}|, \rho_B|a_{\sigma_1\sigma_2}|^3 \ll 1$), the critical temperature for Cooper pairing is

$$T_c = 0.61\, T_F \exp\left[-\frac{\pi/4}{k_F|\bar{a}_{+-}|}\right], \quad (4)$$

where $T_F = \hbar(\mu_+ + \mu_-)R_M/k_B$ is the Fermi temperature with $R_M = m^*_{+-}/\sqrt{m_+ m_-}$. The Fermi wavevector, k_F, was taken as the same for both species, so that $\mu_+ + \mu_- = (m_\pm/m^*_{+-})\mu_\pm$ and $T_F = T_F^{(+)}\sqrt{m_+/m_-}$.

Continuing to focus on ^{87}Rb–^{40}K, $N_B = 1.3 \times 10^6$ BEC atoms in a trap with $\omega_r = 100 \times 2\pi$ Hz and $\omega_a = 10 \times 2\pi$ Hz yield a boson density $\rho_B = 5 \times 10^{14}$ cm^{-3}. A modest number of fermions, $N_F = 5 \times 10^3$, means that the loss of condensate atoms in molecule formation can be neglected, the condensate will absorb any heat created by pairing-induced holes in the atomic Fermi sea [62], and collapse instabilities (Modugno et al. [55]) are avoided. Presuming that fractional STIRAP converts roughly 18% of the initial Fermi atoms into molecules (see Fig. 1), and that the fermions share the same (mass-adjusted) trap, then $\rho_\pm = 1.1 \times 10^{12}$ cm^{-3}, and the requirement of equal Fermi wavenumbers for the atoms and molecules is met. For the given parameters, the size of the BEC is roughly an order of magnitude larger than the Fermi clouds, so that overlap should not be an issue.

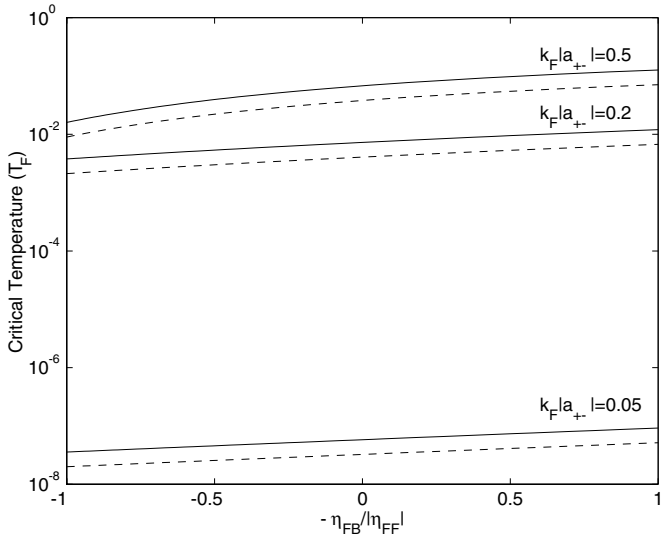

Fig. 4. Critical temperature for the superfluid transition to anomalous atom-molecule pairs as a function of fermion-boson fluctuation strength. Calculations are for weak ($k_F|a_{+-}| = 0.05$), marginally weak (0.2), and marginally strong (0.5) interactions. The solid (dashed) curve gives the critical temperature in units of the effective (atomic) Fermi temperature.

Figure 4 summarizes our investigations. Under the above trap conditions, and for $|a_{+-}| = a_{00} = 5.29$ nm, we find the weakness parameter $k_F|a_{+-}| = 0.05$ and an unreasonably low critical temperature. Nevertheless, if the trap is modified to be anharmonic, then tighter confinement ultimately means a diluteness parameter on the order of $k_F|a_{+-}| = 0.2$ [49], and the situation is improved but still out of reach of current technology ($T \sim 0.05T_F$ [22]). The only chance appears to be for a tight trap and a large scattering length, $k_F|a_{+-}| = 0.5$; however, the theory is at best marginally applicable in this regime and, besides, some other process (e.g., three-body recombination) would probably win out before superfluidity could set-in to such a system.

4 Crossover from a BEC of trimers to atom-dimer Cooper pairs

While possible in principle, accessible atom-molecule superfluidity means that next-generation technology must shed *another* order of magnitude in temperature from the first generation [74], or that a system with an attractive *and* a strong-but-not-too-strong interaction will be found. Our opinion is that it will be worthwhile to look for other ways of driving atom-molecule Cooper pairs, such as a photoassociation or Feshbach resonance.

We therefore model an ideal degenerate mixture of fermionic atoms and dimer molecules coupled by either a Feshbach or photoassociation resonance to bound trimer molecules. An ideal gas is chosen mainly because off-resonant particle-particle interactions are generally too weak for practical purposes. The initial fermionic atom-dimer state could be prepared using a Raman scheme for photoassociating a degenerate Bose-Fermi mixture of atoms (as in the previous section), and selectively removing the leftover bosons. The atom-dimer $\leftrightarrow$ trimer resonance is expectedly well resolved so that, once the initial atom-dimer state has been created, transitions involving three free atoms are avoided. In contrast to the all-boson case [75], ultracold transitions that involve a free bosonic atom are Pauli blocked, i.e., the two identical fermionic constituents of a given trimer may not form a bound state.

In second-quantization parley, a particle of mass m_σ and momentum $\hbar\mathbf{k}$ is described by the creation operator $a_{\mathbf{k},\sigma}$. The Greek index corresponds mnemonically to the number of constituent atoms a given particle contains: 3 for bosonic trimers, 2 for fermionic dimers, and 1 for fermionic atoms. All operators obey their (anti)commutation relations. The microscopic Hamiltonian for such a freely-ideal system is written:

$$\frac{H}{\hbar} = \sum_{\mathbf{k},\sigma} \left[(\epsilon_{k,\sigma} - \mu_\sigma)\, a^\dagger_{\mathbf{k},\sigma} a_{\mathbf{k},\sigma} \right] - \frac{\mathcal{K}}{\sqrt{V}} \sum_{\mathbf{k},\mathbf{k}'} \left(a^\dagger_{\mathbf{k}+\mathbf{k}',3} a_{\mathbf{k},1} a_{\mathbf{k}',2} + \text{H.c.} \right). \quad (5)$$

The free-particle energy is defined by $\hbar\epsilon_{k,\sigma} = \hbar^2k^2/2m_\sigma$, and the chemical potential by $\hbar\mu_\sigma$. In particular, the molecular chemical potential is defined by $\mu_3 = 2\mu - \delta_0$, where the bare detuning δ_0 is a measure of the binding energy of the trimer with $\delta_0 > 0$ taken as above threshold. The (mode-independent) atom-molecule coupling is $\mathcal{K}$, and V is the quantization volume.

The key realization is how to cast the Hamiltonian (5) into a readily diagonalized form. Consider a *time-dependent* unitary transformation, which leaves the physics unchanged providing $H \to U^\dagger H U - iU^\dagger \partial_t U$. Given the generator $U = \Pi_{\mathbf{k},\sigma} \exp[-it u_{\mathbf{k},\sigma} a^\dagger_{\mathbf{k},\sigma} a_{\mathbf{k},\sigma}]$, then $u_{\mathbf{k},3} = u_{\mathbf{k},1} + u_{\mathbf{k},2}$ implies $[H, U] = 0$ and, thus, $H \to H - iU^\dagger \partial_t U$. Appropriately armed, apply the unitary transformation specified by $u_{\mathbf{k},1(2)} = [\epsilon_{k,1(2)} - \epsilon_{k,2(1)}]/2$, which conveniently corresponds to the special case $u_{\mathbf{k},3} = 0$, leaving the trimer term unchanged. The new Hamiltonian reads:

$$\frac{H}{\hbar} = \sum_{\mathbf{k}} \left[(\epsilon_{k,3} + \delta_0 - 2\mu)\, a^\dagger_{\mathbf{k},3} a_{\mathbf{k},3} + (\varepsilon_k - \mu) a^\dagger_{\mathbf{k},\sigma} a_{\mathbf{k},\sigma} \right] - \frac{\mathcal{K}}{\sqrt{V}} \sum_{\mathbf{k},\mathbf{k}'} \left(a^\dagger_{\mathbf{k}+\mathbf{k}',3} a_{\mathbf{k},1} a_{\mathbf{k}',2} + \text{H.c.} \right), \quad (6)$$

where the reduced free-particle energy is $\hbar\varepsilon_k = \hbar^2k^2/4m^*$, with $1/m^* = 1/m_1 + 1/m_2$. Also, chemical equilibrium has been incorporated as $2\mu = \mu_1 + \mu_2$. We may now make a transformation to a dressed basis:

$$\begin{pmatrix} \alpha_{\mathbf{k},1} \\ \alpha^\dagger_{-\mathbf{k},2} \end{pmatrix} = \begin{pmatrix} \cos\theta_k & -e^{i\varphi}\sin\theta_k \\ e^{-i\varphi}\sin\theta_k & \cos\theta_k \end{pmatrix} \begin{pmatrix} a_{\mathbf{k},1} \\ a^\dagger_{-\mathbf{k},2} \end{pmatrix}, \quad (7a)$$

$$\alpha_{\mathbf{k},3} = a_{\mathbf{k},3} + \sqrt{V}\Phi\delta_{\mathbf{k},0}, \quad (7b)$$

where $\delta_{\mathbf{k},0}$ is the Kronecker delta-function,

$$\frac{H}{\hbar} = (\delta_0 - 2\mu)\, V|\Phi|^2 + \sum_{\mathbf{k}} (\epsilon_{k,3} + \delta_0 - 2\mu)\, \alpha^\dagger_{\mathbf{k},3}\alpha_{\mathbf{k},3} + \sum_{\mathbf{k}} \left[(\varepsilon_k - \mu) + \omega_k \left(\alpha^\dagger_{\mathbf{k},1}\alpha_{\mathbf{k},1} + \alpha^\dagger_{\mathbf{k},2}\alpha_{\mathbf{k},2} - 1\right)\right]. \tag{8}$$

The condensate mean-field is $\langle a_{0,3}\rangle/\sqrt{V} = e^{i\varphi}|\Phi|$, the mixing angle is $\tan 2\theta_k = |\Phi|\mathcal{K}/(\varepsilon_k - \mu)$, the quasiparticle frequency is $\omega_k^2 = (\varepsilon_k - \mu)^2 + |\Delta|^2$, and the gap is $|\Delta| = \mathcal{K}|\Phi|$. The Hamiltonian (5) is now lowest-order diagonal.

Consider the mean-field Heisenberg equations for the bosonic operator $a_{0,3}$ and the anomalous-pair-correlation operator $C_{\mathbf{k}} = a_{\mathbf{k},1}a_{-\mathbf{k},2}$ (*sans* chemical potential and collective enhancement):

$$i\frac{d}{dt}\langle a_{0,3}\rangle = \delta_0\langle a_{0,3}\rangle - \frac{\mathcal{K}}{\sqrt{V}}\sum_{\mathbf{k}}\langle C_{\mathbf{k}}\rangle, \tag{9a}$$

$$i\frac{d}{dt}\langle C_{\mathbf{k}}\rangle = 2\varepsilon_k\langle C_{\mathbf{k}}\rangle - \frac{\mathcal{K}}{\sqrt{V}}\langle a_{0,3}\rangle. \tag{9b}$$

Below threshold, simple Fourier analysis delivers the binding energy, $\hbar\omega_B < 0$, of the Bose-condensed trimers:

$$\omega_B - \delta_0 - \Sigma(\omega_B) + i\eta = 0, \tag{10}$$

where $\Sigma(\omega_B)$ is the self-energy of the Bose molecules and $\eta = 0^+$. Incidentally, we show elsewhere [30] that the real poles of equation (10) fit the Regal et al. [22] data for the binding energy of $^{40}K_2$ molecules, and similar measurements for a system of trimers would uniquely determine the parameters of the present theory. On the other side of threshold, the critical temperature for the transition to effectively all superfluid atom-dimer pairs is derived from equation (9a):

$$T_c/T_F \simeq \exp\left(-\frac{\pi/4}{k_F|a_R|}\right). \tag{11}$$

Here the resonant atom-dimer scattering length is $a_R = -(4\pi m^*/\hbar)\mathcal{K}^2/\delta_0$. Also, we have taken a single Fermi wavevector, k_F, for the atoms and the dimers, i.e., $\mu_1 + \mu_2 = \mu_{1(2)}m_{1(2)}/m^*$; assuming that the particles see the same trap, adjusted for mass differences, equal Fermi wavevectors are realized if the number of atoms and dimers satisfy $N_2/N_1 = (m_1/m_2)^{3/2}$. The effective Fermi temperature is $k_B T_F/\hbar = (\mu_1 + \mu_2)m^*/\sqrt{m_1 m_2}$, or $T_F = T_F^{(1)}\sqrt{m_1/m_2}$. Last but not least, it is easy to show $|\Delta| \propto T_c$, so that $|\Phi| \propto \exp(-\pi/4k_F|a_R|)$, and the trimer part of the dressed BEC-pair admixture becomes larger near threshold (increasing $|a_R|$), as expected.

Whereas the below-threshold regime of a trimer condensate is no doubt of interest ($a_R > 0$), both as a precursor to fermionic superfluidity and in its own right, we keep our focus on attractive systems. The strongly interacting regime is defined by $k_F|a_R| \sim 1$, indicating a transition

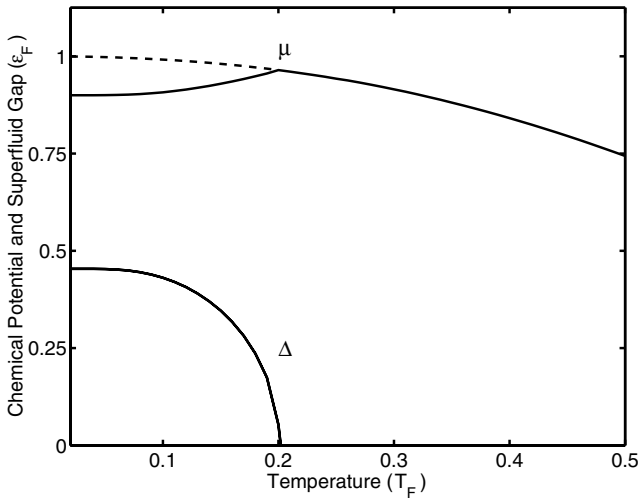

Fig. 5. Onset of the superfluid transition to a Bose-Einstein condensate of trimers dressed by anomalous atom-dimer pairs. The non-zero superfluid gap clearly lowers the system energy compared to the normal state (dashed curve). When the detuning is large and positive ($\delta_2 \gg \sqrt{\rho}\mathcal{K}_2$), such as here, the system is mostly rogue pairs with a negligible fraction of trimers.

to predominantly atom-dimer Cooper pairs at the critical temperature $T_c \sim 0.45T_F$. Using $m_3 = m_1 + 2m_1$ as an example, which is akin to a system of ^{6}Li atoms and ^{7}Li–^{6}Li dimers, the required dimer-atom fraction is $N_2/N_1 = 0.31$, the ratio of the effective and atomic Fermi temperatures $T_F = 0.7T_F^{(1)}$, and the critical temperature $T_c \sim 0.3T_F^{(1)}$. Although equation (11) is of dubious validity for $T_c \lesssim T_F$, it confirms that resonant association should in principle drive superfluid pairing between atoms and dimer molecules at transition temperatures within reach of present ultracold technology.

To rigorously identify the critical temperature for the superfluid transition, it is necessary to go beyond the effective atom-dimer theory, and explicitly include the bosonic molecular state. Continuing to focus on a system of ^{6}Li atoms and ^{7}Li–^{6}Li dimers, we return to the Hamiltonian (8) and set $\epsilon_{k,3} \approx \frac{1}{2}\varepsilon_k$. We also introduce a second molecular state, which can arise because large detuning from one state brings the system into the neighborhood of another bound state, or because of the presence of a scattering resonance. The Hamiltonian (8) is adapted simply by making the substitution $\delta_0 \to \delta_{0,l}$ ($\Phi \to \Phi_l$, $\mathcal{K} \to \mathcal{K}_l$), and summing over the index l; also, the gap becomes $|\Delta| = \mathcal{K}_1|\Phi_1| + \mathcal{K}_2|\Phi_2|$. Here $\mathcal{K}_2 \gtrsim \mathcal{K}_1$, and the system is tuned between the two levels so that $\delta_2 > 0$ and $\delta_1 < 0$.

An algebraic system can be derived [76] from the so-called grand partition function, $\Xi = \mathrm{Tr}\exp(-\beta H)$, which we solve numerically for the chemical potential as a function of temperature, which in turn should display a characteristic cusp at the onset of superfluidity. Physically, a cusp arises because the superfluid BEC-pair dressed state is lower in energy than the normal state, implying the concurrent appearance of a non-zero gap, as shown in Figure 5. For positive detunings large compared to the

collective-enhanced coupling ($\delta_2 \gg \sqrt{\rho}\mathcal{K}_2$ with $\delta_1 \approx -\delta_2$), the effective atom-dimer theory (11) with $k_F|a_R| = 1/2$ is an excellent working approximation. Also, the fraction of trimer is puny ($\sim 10^{-7}$), as per the large detuning. Any s-wave collisional interactions are negligible compared to the detuning, justifying the ideal-gas assumption. The trap, albeit omitted, should actually favor the occurrence of superfluid pairing [48].

Dimer molecules created near a Feshbach resonance are highly vibrationally excited and, thus, characteristically long-range (Köhler et al. [15]). Fermion-composite dimers are consequently long-lived due to Pauli-suppressed vibrational relaxation, and there is no reason to expect otherwise from Feshbach trimer states. In photoassociation, a two-color Raman scheme is required to avoid spontaneous decay: a laser couples the atoms to an electronically-excited intermediate trimer state, a second laser couples the system to a ground-electronic trimer manifold, and the intermediate trimer state is well-detuned. Long-range states are available with photoassociation, although a Raman scheme also allows access to stable lower-lying vibrational levels, which are much smaller and less understood. Nevertheless, the molecular fraction is negligibly small when the system is well above the appropriate threshold, diminishing the chance for spontaneous for decay of any kind.

5 Summary and outlook

In conclusion, we highlight that the term $\Omega = \sqrt{N}\,\kappa$ was previously referred to as the Bose-enhanced free-bound coupling; 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. Consequently, we find that collective Rabi oscillations and rapid adiabatic are feasible in principle, and that the latter should also be possible in practice. Similarly, stimulated Raman adiabatic passage from a degenerate Bose-Fermi mixture to Fermi molecules should be possible, even in the presence of collisions. On the other hand, while possible in principle, achieving off-resonant atom-molecule superfluidity with current ultracold technology means that a system with an attractive *and* a strong-but-not-too-strong interaction must be found. Hence, we suggest searching for an association resonance that combines atoms and diatomic molecules into triatomic molecules, which leads to a crossover from a BEC of trimers to atom-dimer Cooper pairs at a temperature reachable with current techniques.

Each of the above results can be reinvestigated to determine the effect of long-range dipolar interactions. Off hand, Rabi-like oscillations are not likely to bear much fruit: the usual s-wave collisions lead to mean-field shifts that make exact resonance, and thus large-amplitude Rabi oscillations, difficult to achieve; the long-range dipolar interaction will most probably make this difficult situation even worse. But, we expect that rapid adiabatic passage survives the inclusion of particle interactions, as in the all-boson case [63], and the addition of a dipolar interactions for the molecules could sideline this method for creating heteronuclear molecules. Moreover, STIRAP is highly sensitive to particle-particle interactions [69], and it is possible that inclusion of the long-range dipolar interaction could moot this technique for anything other than simple heteronuclear systems (e.g., ^{6}Li–^{7}Li). Besides the formation of heteronuclear molecules with an association resonance, the role of dipolar interactions in atom-molecule Cooper pairing could also be investigated, which raises an interest because the dipolar molecule will induce a dipole moment in the atom (as opposed to pairing between two permanent *or* induced dipolar particles [77]).

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