Feshbach-Resonant Interactions in ^{40}K and ^{6}Li Degenerate Fermi Gases

Matt Mackie[1] and Jyrki Piilo[2]

[1]*QUANTOP-Danish National Research Foundation Center for Quantum Optics, Department of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus C, Denmark*

[2]*School of Pure and Applied Physics, University of KwaZulu-Natal, Durban 4041, South Africa*

(Received 1 November 2004; published 15 February 2005)

We theoretically examine a system of Fermi degenerate atoms coupled to bosonic molecules by a Feshbach resonance, focusing on the superfluid transition to a molecular Bose-Einstein condensate dressed by Cooper pairs of atoms. This problem raises interest because it is unclear at present whether bimodal density distributions observed recently in ^{40}K and ^{6}Li are due to a condensate of bosonic molecules or fermionic atom pairs. As opposed to ^{40}K, we find that any measurable fraction of above-threshold bosonic molecules is necessarily absent for the ^{6}Li system in question, which strongly implicates Cooper pairs as the culprit behind its bimodal distributions.

DOI: 10.1103/PhysRevLett.94.060403 PACS numbers: 03.75.Ss

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] and condensate [3] molecules have been created via magnetoassociation of a Fermi gas of atoms, in the course of efforts to create superfluid Cooper-paired atoms [4,5] (see also Ref. [6]). In particular, for a magnetic field tuned above the two-body threshold for molecule formation, the observation of a bimodal density distribution for a system of ^{40}K atoms was attributed to the existence of a Bose-Einstein condensate of fermionic Cooper pairs [4]. Nevertheless, a lone theoretical analysis suggests that the ^{40}K data [4] can be understood as a Bose-Einstein condensate of molecules (MBEC), since the presence of the Fermi sea shifts the threshold for molecular formation to the point where the molecular binding energy is equal to twice the Fermi energy [7], an interpretation that has been bolstered, though not confirmed, by observations of bimodal distributions in ^{6}Li atoms [5]. The purpose of this Letter is to demonstrate that significant fractions of above-threshold molecular condensates are absent only when the atom-molecule coupling is much larger than the Fermi energy. In a surprise reversal, our results point to interpretations of a molecular and fermionic Bose-Einstein condensates, respectively, instead of fermionic [4] and molecular [5] condensates.

The mean-field theory of magnetoassociation of a Fermi gas of atoms leads to two types of instabilities against molecule formation. The first one is a dynamical instability, whereby the larger state space of the molecules, owing somewhat to Pauli blocking, leaves the atoms prone to spontaneous association into molecules [8]. Here we focus on the thermodynamic instability of a Fermi sea against the formation of Cooper pairs [9], a trait of superconductors whose analog is passed on to Feshbach-resonant superfluids [10]. A thermodynamical instability occurs because pairing lowers the energy, similar to Fig. 1, and so coupling to a reservoir with a low enough temperature leaves the system prone to pairing. The question is what role molecules play in this process.

Our answer is outlined as follows. After introducing the model [11], we show that weak (strong) coupling gives a large (negligible) fraction of above-threshold molecules and that, although contrary to two-body physics, this result makes perfect sense in terms of our previous [12,13] boson results. In particular, for atom-molecule couplings much larger than the Fermi energy, dissociation to fermionic pairs should dominate the creation of bosonic molecules. We then find that bosonic molecules can be ruled out for the observations in Ref. [5], but not for those in Ref. [4]. Before concluding, we contrast our results with the recent related theories [7,14–17].

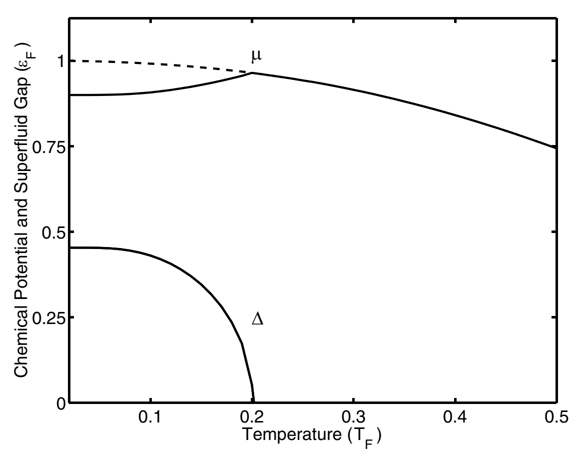

FIG. 1. An example of the onset of the superfluid transition to a Bose-Einstein condensate of molecules dressed by fermionic atom pairs. The appearance of a nonzero superfluid gap (Δ) lowers the chemical potential (μ) compared to the normal state (dashed curve), signaling the onset of the superfluid regime. As it happens, the coupling constant and the detuning in this example are chosen to be exceptionally large (see text), similar to recent experiments [5].

Ideal gas model.—We model an ideal two-component gas of fermionic atoms coupled by a Feshbach resonance to bosonic molecules. An ideal gas is chosen for simplicity and is justified by a collisional interactions strength that is negligible compared to the atom-molecule coupling (see next-to-last section). In the language of second quantization, an atom of mass m and momentum $\hbar\mathbf{k}$ is described by the annihilation operator $a_{\mathbf{k},1(2)}$, 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:

$$\frac{H}{\hbar} = \sum_{\mathbf{k}}\left[(\epsilon_k - \mu)a^\dagger_{\mathbf{k},\sigma}a_{\mathbf{k},\sigma} + \left(\frac{1}{2}\epsilon_k + \delta_0 - \mu_{\rm mol}\right)b^\dagger_{\mathbf{k}}b_{\mathbf{k}}\right] - \frac{\mathcal{K}}{\sqrt{V}}\sum_{\mathbf{k},\mathbf{k}'}(b^\dagger_{\mathbf{k}+\mathbf{k}'}a_{\mathbf{k},1}a_{\mathbf{k}',2} + \text{H.c.}), \tag{1}$$

where repeated greek indices imply a summation ($\sigma = 1, 2$). The free-particle energy is $\hbar\epsilon_k = \hbar^2k^2/2m$, the atom (molecule) chemical potential is $\hbar\mu_{\sigma(\rm mol)}$, and the bare detuning δ_0 is a measure of the binding energy of the molecule ($\delta_0 > 0$ is taken as above threshold), the mode-independent atom-molecule coupling is $\mathcal{K}$, and V is the quantization volume. We have already imposed the ideal conditions for superfluidity with $\mu_1 = \mu_2 = \mu$, and now we impose chemical equilibrium between the atoms and molecules with $\mu_{\rm mol} = \mu_1 + \mu_2 = 2\mu$. Diagonalization of the Hamiltonian (1) is achieved by the standard transformation to a dressed basis [11]:

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

$$\beta_{\mathbf{k}} = b_{\mathbf{k}} - \sqrt{V}\Phi\delta_{\mathbf{k},0}, \tag{2b}$$

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

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

The condensate mean field is $\langle b_0\rangle/\sqrt{V} = e^{i\varphi}|\Phi|$, the mixing angle is $\tan 2\theta_k = |\Delta|/(\epsilon_k - \mu)$, the eigenfrequencies are $\omega_k^2 = (\epsilon_k - \mu)^2 + |\Delta|^2$, and the gap is $|\Delta| = |\Phi|\mathcal{K}$.

To determine the thermodynamic ground state, we first calculate the pressure from the partition function $\Xi = \text{Tr}\exp(-\beta H)$, which is then extremized with respect to the molecular amplitude and, in turn, the chemical potential, yielding [11]

$$(\delta - 2\mu) = \Sigma(0) + \frac{\mathcal{K}^2}{2V}\sum_{\mathbf{k}}\frac{1}{\omega_k}\tanh\frac{1}{2}\beta\omega_k. \tag{4a}$$

$$\rho = 2|\Phi|^2 + \frac{2}{V}\sum_{\mathbf{k}}\frac{1}{\exp[\beta(\frac{1}{2}\epsilon_k + \delta - 2\mu)] - 1} + \frac{1}{V}\sum_{\mathbf{k}}\frac{\omega_k + \mu - \epsilon_k + (\omega_k - \mu + \epsilon_k)\exp(-\beta\omega_k)}{\omega_k[1 + \exp(-\beta\omega_k)]}. \tag{4b}$$

Renormalization is via the resonant self-energy $\Sigma(0)$ [13], meaning the physical detuning δ replaces the bare δ_0.

Weak versus strong coupling.—Solving the algebraic system (4) self-consistently determines the chemical potential as a function of temperature. Intuitively the appearance of a nonzero gap lowers the chemical potential of the superfluid MBEC-pair dressed state compared to the normal state, which allows us to confirm that, for a given temperature, the system is in the superfluid regime, cf. Fig. 1. Along with the conservation of particle number, Figs. 2(a) and 2(b) indicate a ground state that is a molecular condensate dressed by dissociated fermionic pairs, an admixture adjustable according to the detuning from threshold. In particular, for an atom-molecule coupling that is weak compared to the Fermi energy, $\Omega = \sqrt{\rho}\mathcal{K} \lesssim \epsilon_F$ [where $\epsilon_F = \hbar(3\pi^2\rho)^{2/3}/2m$], we find a large fraction of above-threshold molecular condensate [Fig. 2(a)], whereas $\Omega \gg \epsilon_F$ leads to an above-resonance system that is predominantly fermionic pairs [Fig. 2(b)].

We have confirmed that for strong coupling the fraction of molecular condensate remains negligible ($\sim 10^{-3}$) for $\delta \sim \epsilon_F$; i.e., the absence of a large near-threshold fraction is not the result of choosing Ω as the frequency scale; similarly, having properly renormalized the detuning, it is not due to any spurious shift of resonance threshold. Additionally, the trap, albeit omitted, can actually favor the occurrence of superfluid pairing [18].

Below threshold ($\delta < 0$), Fourier analysis delivers the binding energy, $\hbar\omega_B < 0$, of the Bose-condensed molecules [8,13]: $\omega_B - \delta - \Sigma'(\omega_B) + i\eta = 0$, where $\Sigma'(\omega_B)$ is the finite self-energy of the Bose molecules and $\eta = 0^+$. Above the two-body threshold ($\delta > 0$) gives an imaginary ω_B, and the bound state ceases to exist; nevertheless, Fig. 2(a) shows a large fraction of molecular condensate, which drops off for increasing coupling strength as per Fig. 2(b). These apparently contradictory results are in fact consistent with the dynamical studies of rapid adiabatic passage in bosons [12,13]: just above threshold, the coupled atom-molecule system can have a significant fraction of molecular condensate [12], but "rogue" dissociation to atom pairs with equal-and-opposite momentum is expected to dominate the formation of molecules for $\Omega \gg \hbar\rho^{2/3}/m \approx \epsilon_F$ [12,13]. Physically, the rate for converting atoms into molecules is $\sim\Omega$, whereas the rate for rogue

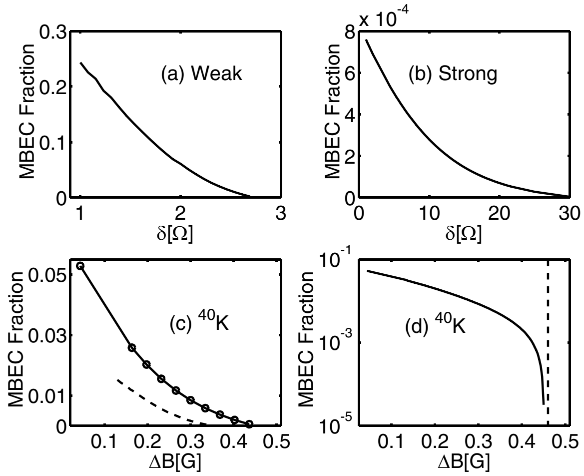

FIG. 2. Molecular condensate fraction as a function of the above-threshold detuning. For $T = 0.05T_F < T_C$, where T_C is the critical temperature for the superfluid transition, weak coupling ($\Omega = \epsilon_F$) finds a large fraction of above-threshold molecular condensate (a), whereas a strong coupling ($\Omega = 35\epsilon_F$) finds a largely absent MBEC (b), independent of the above-threshold detuning. In (c), the solid line is for $T = 0.08T_F < T_C$ and $\Omega = 4\epsilon_F$, and the dashed line is the weak result from (a) scaled by $1/4^2$ [21]. The open circles are used to emphasize how only the first point is taken from the region where the ideal gas approximation may break down. The actual results are markedly better than the scaling, and in agreement with observation [4] except for a shift of about 0.2 G. In (d), the superfluid ^{40}K phase boundary is evident in a departure from exponential behavior.

magnetodissociation is $\Gamma_0 \propto \Omega^2$ [19]; hence, for a strong enough coupling, rogue magnetodissociation to fermionic pairs of atoms with equal-and-opposite momentum will dominate the formation of molecular condensate.

Recent experiments and theory.—The significance of the above results is seen by comparison with experiments in ^{40}K [4] and ^{6}Li [5] systems. Detunings are converted into magnetic fields according to $\Delta B = \hbar\delta/\Delta_\mu$, where the difference in magnetic moments between the atom pair and a molecule is Δ_μ, and where $\Delta B = B - B_0$ (with B_0 the magnetic-field position of resonance).

For a ^{40}K gas of density $\rho = 2 \times 10^{13}\mathrm{cm}^{-3}$, the coupling strength is [8,20] $\Omega \approx 4\epsilon_F$; the difference in magnetic moments is $\Delta_\mu \approx 0.19\mu_0$ [8] (with μ_0 the Bohr magneton). Offhand, we are tempted to scale [21] the results of Fig. 2(a), but the result is not encouraging [Fig. 2(c), dashed line]; nevertheless, a full recalculation [Fig. 2(c), solid line and open circles] of the solution to Eqs. (4) for $\Omega = 4\epsilon_F$ yields results that, except for a roughly 0.2 G shift, agree embarrassingly well with the measured [4] bimodal distributions (not shown). The 0.2 G disagreement is understood more clearly given Fig. 2(d), where the superfluid phase boundary in detuning, given roughly by the dashed line, is marked by a clear departure from exponential behavior. Overall, it seems that we agree with Ref. [7] that bosonic molecules cannot be ruled out, by two-body physics or otherwise, as the culprit responsible for the condensate footprints in ^{40}K [4].

On the other hand, for a ^{6}Li gas of typical density [5], the atom-molecule coupling is [20,22,23] $\Omega = 87\epsilon_F$. Approximating $\Delta_\mu = 2\mu_0$, any MBEC can already be ruled out of experiments [5] based on Fig. 2(b). Even without explicitly accounting for the somewhat larger ^{6}Li coupling constant, Fig. 2(b) would predict a near-unit fraction of fermionic pairs. Since the atom-molecule coupling in Fig. 1 is $\Omega \sim 150\epsilon_F$ and the detuning is $\delta \gg \Omega$, we estimate the critical temperature $T_C \sim 0.2T_F$, which is close to the measured [5] large-detuning critical temperature (all things considered); hence, we expect $T = 0.05T_F$ to be far enough below the critical temperature so that what is not molecular condensate is most likely superfluid fermionic pairs, with only a small thermal fraction. Indeed, whereas initial experiments measure 80% condensate fractions [5], recent measurements are in excess of 90% [23], consistent with expectation.

We pause to briefly justify the ideal gas model. The collisional interaction strength is $\Lambda = 2\pi\hbar\rho a/m$, where a is the off-resonant atomic s-wave scattering length. The ^{40}K and the ^{6}Li scattering length are roughly an order of magnitude apart: $a_{\mathrm{K}} = 176a_0$ [24] and $|a_{\mathrm{Li}}| = 2110a_0$ [22,25], with a_0 the Bohr radius. For a typical density $\rho \sim 10^{13}\mathrm{cm}^{-3}$, it turns out that the magnitude of the collisional coupling strengths, in units of the atom-molecule coupling, is roughly equal: $|\Lambda|/\Omega \approx 10^{-3}$. Collisions should therefore be broadly negligible.

Before closing, it is important to draw contrast with the latest work of others. First, it appears that the stability of the above-threshold molecules is a matter of competition between formation of molecular condensate and the subsequent magnetodissociation to fermionic pairs, as opposed to many-body effects [7] (see also Ref. [14]). Next, although the chemical potential for the near-resonant ^{40}K system is undoubtedly in the so-called universal regime [15], and molecules are not expected to play role in such a case [15], we found herein that molecules cannot be ruled out. Also, whereas a prominent single-channel theory [26], i.e., only atoms and their pairs, no explicit molecules, has shown good agreement with the ^{40}K experiments [16], this theory would presumably deliver similar answers for both experiments [4,5], unlike our theory. Finally, it is entirely possible that the threshold of a strongly coupled system is systematically shifted to negative detunings, as suggested recently [17]—albeit without recourse to the atom-molecule coupling strength, a matter deferred elsewhere.

Summary and conclusions.—We have found that, in a Feshbach-resonant gas of Fermi atoms, significant fractions of above-threshold molecular condensate are absent for atom-molecule couplings that are strong compared to the Fermi energy. While it is perhaps a stretch call the ^{40}K [4] system weak, it is clear that bosonic molecules cannot be ruled out of its bimodal distributions. However, bosonic molecules can be ruled out for the ^{6}Li [5] system, which strongly suggests that Cooper pairing/fermionic condensation has been observed. Our interpretation is that, because strong rogue magnetodissociation favors fermionic pair formation, a condensate of Cooper pairs rather than molecules is formed. These results suggest that, in addition to bosons and fermions, the schism between dynamics and thermodynamics is, at least on some level, artificial. Moreover, debate is currently raging over the necessity of a separate molecular channel in theories of Feshbach-resonant fermionic atoms [27], and our results strongly suggest that molecules play an explicit role in a holistic understanding of experiments.

We kindly thank Michael Budde, Eric Cornell, Jason Ho, Randy Hulet, Nicolai Nygaard, Henk Stoof, and Martin Zwierlein for valuable insight, as well as the Academy of Finland (J. P., Project No. 204777) and the Magnus Ehrnrooth Foundation for support (J. P.).

[1] W. C. Stwalley, Phys. Rev. Lett. **37**, 1628 (1976); E. Tiesinga, B. J. Verhaar, and H. T. C. Stoof, Phys. Rev. A **47**, 4114 (1993).
[2] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature (London) **424**, 47 (2003); K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003); J. Cubizolles *et al.*, *ibid.* **91**, 240401 (2003).
[3] M. Greiner, C. A. Regal, and D. S. Jin, Nature (London) **426**, 537 (2003); S. Jochim *et al.*, Science **302**, 2101 (2003); M. W. Zwierlein *et al.*, Phys. Rev. Lett. **91**, 250401 (2003).
[4] C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett. **92**, 040403 (2004).
[5] M. W. Zwierlein *et al.*, Phys. Rev. Lett. **92**, 120403 (2004).
[6] C. Chin *et al.*, Science **305**, 1128 (2004); J. Kinast *et al.*, Phys. Rev. Lett. **92**, 150402 (2004); T. Bourdel *et al.*, Phys. Rev. Lett. **93**, 050401 (2004).
[7] G. M. Falco and H. T. C. Stoof, Phys. Rev. Lett. **92**, 130401 (2004).
[8] J. Javanainen *et al.*, Phys. Rev. Lett. **92**, 200402 (2004).
[9] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).
[10] 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).
[11] R. Friedberg and T. D. Lee, Phys. Rev. B **40**, 6745 (1989).
[12] J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999).
[13] M. Mackie, K.-A. Suominen, and J. Javanainen, Phys. Rev. Lett. **89**, 180403 (2002).
[14] J. E. Williams, T. Nikuni, N. Nygaard, and C. W. Clark, cond-mat/0403503; J. Phys. B **37**, L351 (2004).
[15] R. B. Diener and T.-L. Ho, cond-mat/0405174.
[16] A. V. Avdeenkov and J. L. Bohn, cond-mat/0404653 [Phys. Rev. A (to be published)].
[17] M. M. Parish, B. Mihaila, E. M. Timmermans, K. B. Blagoev, and P. B. Littlewood, cond-mat/0410131 [Phys. Rev. B (to be published)].
[18] C. P. Search, H. Pu, W. Zhang, B. P. Anderson, and P. Meystre, Phys. Rev. A **65**, 63616 (2002).
[19] The simplest way to infer these rates is by the Heisenberg equations of motion for the Hamiltonian (1).
[20] R. G. Hulet (private communication).
[21] Based loosely on Timmermans *et al.* [10], we find that the molecular fraction $f = 2|\Phi|^2/\rho$ scales as $f/f_0 = (\bar{\Omega}_0/\bar{\Omega})^2$, where $\bar{\Omega} = \Omega/\epsilon_F$.
[22] M. Houbiers, H. T. C. Stoof, W. I. McAlexander, and R. G. Hulet, Phys. Rev. A **57**, R1497 (1998); E. R. I. Abraham *et al.*, *ibid.* **55**, R3299 (1997).
[23] M. Zwierlein (private communication).
[24] J. L. Bohn, Phys. Rev. A **61**, 053409 (2000).
[25] M. Bartenstein *et al.*, cond-mat/0408673.
[26] S. J. J. M. F. Kokkelmans, J. N. Milstein, M. L. Chiofalo, R. Walser, and M. J. Holland, Phys. Rev. A **65**, 053617 (2002).
[27] Proceedings of the KITP Conference: Quantum Gases, Santa Barbara, California, 2004, http://online.kitp.ucsb.edu/online/gases_c04/