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Production of Long-Lived Ultracold Li_2 Molecules from a Fermi Gas

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We create weakly bound Li_2 molecules from a degenerate two component Fermi gas by sweeping a magnetic field across a Feshbach resonance. The atom-molecule transfer efficiency can reach 85% and is studied as a function of magnetic field and initial temperature. The bosonic molecules remain trapped for 0.5 s and their temperature is within a factor of 2 from the Bose-Einstein condensation temperature. A thermodynamical model reproduces qualitatively the experimental findings.

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Feshbach resonances constitute a unique tool to tune the microscopic interactions in ultracold bosonic and fermionic gases [1–3]. These resonances arise when the total energy of a pair of colliding atoms matches the energy of a bound state of another hyperfine manifold, leading to the resonant occupation of this state during the collision. Thus, by means of an external magnetic field, one is able to change the magnitude and sign of the scattering length. In bosonic samples, the collapse of Bose-Einstein condensates (BEC) for negative scattering length [4], soliton formation [5,6], and coherent oscillations between an atomic condensate and molecules [7] have been observed. For fermions with attractive interaction, the superfluid transition temperature is predicted to be maximum near a Feshbach resonance [8–12].

In this Letter, we give another striking example of the control of interactions in a Fermi gas. We perform time-dependent experiments near a Feshbach resonance to produce in a reversible manner ultracold and trapped molecules from a quantum degenerate fermionic ^{6}Li gas. The production efficiency exceeds 80% and the observed molecule lifetime reaches half a second. The phase-space density of these bosonic molecules is on the order of 1, the highest value reported thus far. Using a similar method with fermionic ^{40}K atoms, the JILA group recently reported molecule production with a lifetime of 1 ms, atom to molecule conversion efficiency of 50% and direct measurement of the molecular binding energy [13]. Molecules have also recently been produced from ^{87}Rb and ^{133}Cs condensates [14,15], from a cold ^{133}Cs cloud [16]. Molecule formation has also been achieved through one-photon or two-photon photoassociation but with a considerably lower phase-space density than reported here [17]. Our work paves the way to Bose-Einstein condensation of molecules and to the study of the crossover between the regime of molecular BEC and the regime of superfluid BCS pairing in Fermi systems [8–12].

In ^{6}Li a broad ($\simeq$100 G) Feshbach resonance exists between the two Zeeman sublevels of the hyperfine ground state : $|1/2, -1/2\rangle$ and $|1/2, 1/2\rangle$ at 810 G, see Fig. 1 [18,19]. In a recent study of this Feshbach resonance, we have reported an anomalous negative value for the gas interaction energy between 700 G and 810 G, i.e., below resonance, where the scattering length a is positive [19]. We suggested that this could be due to the presence of weakly bound molecules confined simultaneously with the cloud of ultracold fermions, and a recent theoretical paper explains our results [20].

The method used here to produce molecules is illustrated in Fig. 1 and was suggested for Bose gases in [21–23]. It consists in scanning over a Feshbach resonance from the region of attractive interaction ($a < 0$) to region 2 in Fig. 1, where a is large and positive, and where a weakly bound molecular state exists with energy $E_b = -\hbar^2/ma^2$ (where m is the atomic mass). It is thus energetically favorable to populate this bound state. Having

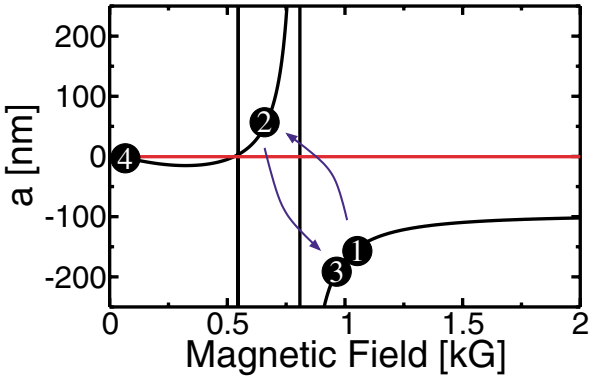

FIG. 1 (color online). Calculated scattering length a versus magnetic field for the ^{6}Li $|F, m_F\rangle = |1/2, 1/2\rangle$, $|1/2, -1/2\rangle$ mixture near the 810 G Feshbach resonance. Scanning over the resonance from 1 to 2 in 50 ms produces cold and trapped molecules with an efficiency up to 85%, resulting in an almost complete disappearance of the atomic signal at 2. Reversing then the scan to the initial position reestablishes the initial atomic signal. In all cases, atoms are detected and imaged at position 4 after abrupt switch-off of the B field.

twice the polarizability of individual atoms, the molecules are expected to be confined in an optical dipole trap with the same oscillation frequencies as atoms and twice the trap depth.

Using the setup described in [19], we prepare $N_1 = N_\uparrow + N_\downarrow = 1.5 \times 10^5$ atoms in a 50(10)% mixture of $|1/2, -1/2\rangle$ and $|1/2, 1/2\rangle$ in a crossed Nd:YAG dipole trap at $B = 1060$ G where the scattering length between $|1/2, -1/2\rangle$ and $|1/2, 1/2\rangle$ is large and negative $a = -150$ nm (position 1 in Fig. 1). For a power of 1.53 W (4.3 W) in the horizontal (vertical) trapping beam propagating along Ox (Oy), oscillation frequencies are $\omega_x/2\pi = 2.9(3)$ kHz, $\omega_y/2\pi = 5.9(5)$ kHz, and $\omega_z/2\pi = 6.5(6)$ kHz. By evaporation in the optical trap, the temperature T of the gas mixture can be tuned between 0.2 and $0.5T_F$, where T_F is the Fermi temperature defined by $T_F = \hbar\bar{\omega}(3N_1)^{1/3}/k_B$ and $\overline{\omega} = (\omega_x\omega_y\omega_z)^{1/3}$.

The Fermi gas quantum degeneracy is measured through analysis of absorption images after abrupt (20 μs) switch-off of the magnetic field and time of flight expansion of the cloud. Free ^{6}Li atoms are detected at zero magnetic field, for which $a \simeq 0$, using laser light tuned to the D2 line, position 4 in Fig. 1. Having prepared the degenerate Fermi gas at position 1, we sweep the magnetic field in 50 ms to position 2 where a is large and positive, the region where a weakly bound molecular state exists. Absorption images indeed reveal that the number of atoms N_2 as counted after B switch-off to position 4 as explained above, has dramatically decreased to $\simeq$20% of the initial number N_1. After letting the system at position 2 for a variable wait time t_w between 0 and 5 s, we sweep the magnetic field back to position 1 in 50 ms and count the number of atoms, N_3, after this round-trip through resonance.

Surprisingly, when position 2 is at 689 G and $t_w = 0$, we find $N_3 \simeq N_1$, indicating that no loss has occurred in 100 ms. This proves that the decrease in detected atom number at position 2 is not due to losses. We are thus led to conclude that (i) at position 2, 80% of the atoms were in a molecular bound state which is not detected using light resonant with the atoms at position 4; (ii) the atom-to-molecule formation process is reversible. $(N_3 - N_2)/2$ represents thus the number of molecules at position 2.

The relative fraction of atoms bound in a molecule, $(N_3 - N_2)/N_3$, and the temperature of the atoms at position 2 and position 3 are plotted in Fig. 2 as a function of the magnetic field at position 2. The initial parameters at 1060 G for this experiment are $T = 4.7$ μK, $T_F = 11$ μK, and $N_1 = 8\,10^4$. The frequencies of the trap are $\omega_x/2\pi = 2.2(2)$ kHz, $\omega_y/2\pi = 4.6(4)$ kHz, $\omega_z/2\pi = 5.1(5)$ kHz. One observes that very few molecules are detected above 0.77 kG. When decreasing the magnetic field, the fraction of molecules increases up to 60%. Below 650 G, losses become important and the molecular fraction decreases, an effect that we will study below.

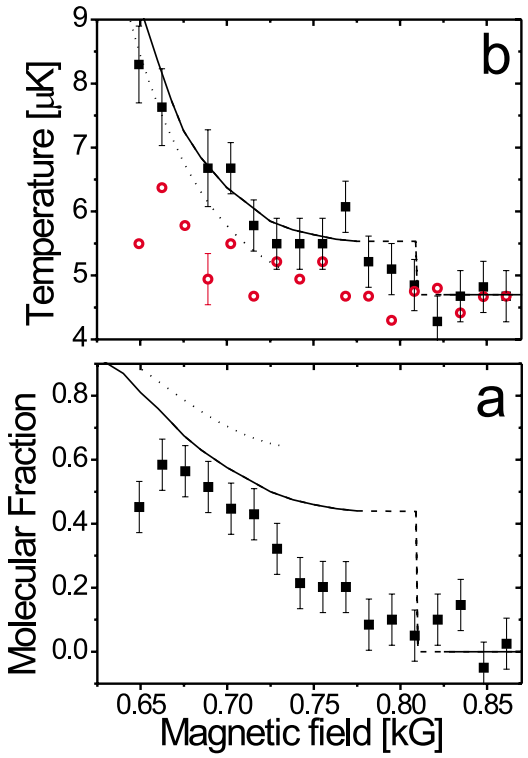

FIG. 2 (color online). (a) Fraction of atoms in the molecular bound state, $(N_3 - N_2)/N_3$, versus magnetic field at position 2 in Fig. 1. (b) Corresponding atom temperature at position 2 (squares) and position 3 (circles). Solid (dotted) line corresponds to the noninteracting (interacting) thermodynamical model described in the text.

A key parameter in our detection scheme is the switch-off time (20 μs) of the Feshbach magnetic field from position 2. Indeed, the molecules may or may not dissociate in this process. Molecules will dissociate if the relative rate of change of the binding energy $dE_b/E_b dt$ is greater than the oscillation pulsation $E_b/\hbar$. So, the dissociation of molecules is governed by the parameter $\alpha = \hbar dE_b/E_b^2 dt$. If $\alpha \gg 1$, they dissociate and appear as free atoms in the absorption image. Our detection scheme is unable to prove molecule formation. On the contrary, if the binding energy is sufficiently large at position 2 ($\alpha \ll 1$), the molecular state follows adiabatically the change of B to a deeper bound state which is off-resonant with the detection laser. In this case, $N_3 > N_2$ is the signature of molecule formation.

For $B = 700$ G, one has $\alpha \simeq 1$. Reducing the rate dB/dt by a factor of $\simeq 500$, the threshold for molecule detection is shifted to 800 G, as expected from the expression of α and the position of the Feshbach resonance at 810 G [19]. As a second consequence of the role of the switch-off time, we attribute the decrease of the molecular fraction above 720 G in Fig. 2(a) to the crossover between the adiabatic and nonadiabatic regimes.

The temperature of the atoms at position 2 and position 3 is plotted in Fig. 2(b). A decrease of the magnetic field

increases the binding energy of the molecules. Therefore, the molecular fraction increases and so does the temperature. However, sweeping back the magnetic field to the initial position, the temperature returns close to its initial value, demonstrating the reversibility of the molecular formation process. According to [24], in our experiment three-body recombination to the weakly bound state occurs at a rate greater than 10^4 Hz (for $B > 665$ G) and is therefore responsible for molecule formation. Moreover, the atom-atom, atom-molecule, and molecule-molecule [25] collision rates are also large compared to the rate at which the resonance is scanned. Consequently, the system remains close to thermal equilibrium. During the formation of a molecule in a three-body recombination process, the binding energy is converted into kinetic energy. Therefore the increase of temperature in Fig. 2(b), as a decreases, is expected.

Interestingly, the efficiency of molecule formation strongly depends on the trap depth and reaches values up to 80 % in Fig. 3, the highest efficiency reported thus far. The trap depth is slowly (800 ms) reduced at $B = 1060$ G, before the magnetic field sweep through resonance. The gas temperature and the Fermi temperature decrease by evaporation and adiabatic cooling due to the reduction of the trap oscillation frequencies. At 689 G, $a = 78$ nm, $E_b/k_B = 12$ μK. When reducing the trap depth, T and T_F become small compared to E_b and the molecular fraction increases towards 1. Other experiments show that both evaporation (reduction of the ratio T/T_F), and adiabatic cooling (reduction of T and T_F by the same factor) effectively lead to an increase of the molecular fraction. For instance, at constant $T_F = 11$ μK, as the temperature is decreased from $0.5T_F$ to $0.3T_F$, we find that the molecular fraction at 689 G increases from 40% to 65%. Similarly, at constant $T/T_F = 0.3$, as T varies from 4.5 μK to 1.8 μK, it increases from 50% to 85%.

Our results can be understood within a thermodynamical model assuming that atoms and molecules are in equilibrium in a trap during the magnetic field sweep [20]. We assume an adiabatic sweep, entropy is then conserved and calculated for the initial situation where no molecules are present at $B = 1060$ G. The atom-atom, atom-molecule, and molecule-molecule interactions were either neglected (solid line in Fig. 2), or included via a mean field contribution proportional to a [24,25], in the regime where it is allowed (dotted line). The molecular fraction is then determined by a set of two parameters T/T_F and E_b/T_F. As the binding energy gets more negative, the fraction of molecules increases since the molecular state becomes energetically more favorable. Moreover, the temperature increases as each molecule formation event transfers the binding energy into kinetic energy. Both models agree fairly well with our data in the region where both the detection of molecules is efficient and losses are unimportant ($660 \lesssim B \lesssim 720$ G). In the interacting case, a perfect agreement is achieved if we allow for a 10% mismatch in the quality of the spin mixture. In the strongly interacting region where $na^3 \geq 1$, the noninteracting model is shown with a dashed line because we do not expect our model of separated gases of fermionic atoms and bosonic molecules to be valid. Those models also account qualitatively for the increase in the molecular fraction with the evaporation in Fig. 3. Indeed, as the trap depth is lowered, T decreases, the ratio E_b/T increases, and the bound state will then have a higher occupancy.

In order to investigate prospects for the formation of a molecular BEC, two questions immediately arise: What are the lifetime and degeneracy parameter of the produced molecular cloud? The decay of the molecules in the optical trap is found by measuring the quantity $(N_3 - N_2)/2$ as a function of the wait time t_w at position 2 before returning to 3 for the detection process. Figure 4 presents the number of remaining molecules as a function of the time elapsed at point 2 in a trap with frequencies $\omega_x/2\pi = 0.95(10)$ kHz, $\omega_y/2\pi = 2.0(2)$ kHz, $\omega_z/2\pi = 2.2(2)$ kHz. In Figs. 4(a) and 4(b), the initial temperatures at position 1 are the same, 1.1 μK $= 0.3T_F$. Figure 4(a) corresponds to $B = 689$ G, $a = 78$ nm, $E_b/k_B = 12$ μK,

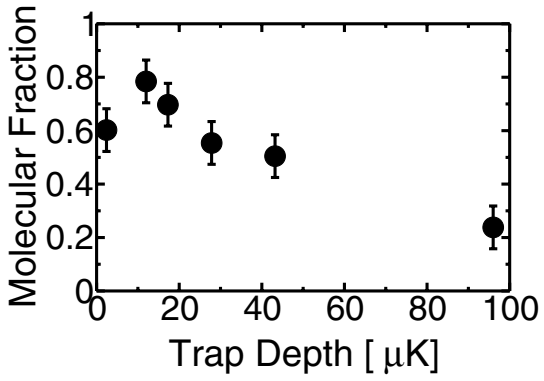

FIG. 3. Fraction of atoms bound in a molecule as a function of the depth of the dipole trap for $B = 689$ G, $a = 78$ nm, $E_b = 12$ μK.

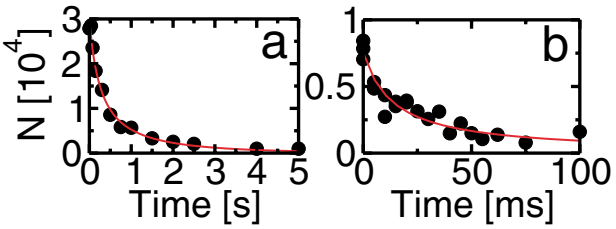

FIG. 4 (color online). Decay of Li_2 molecules in the optical trap for two values of the magnetic field: (a) $B = 689$ G, $a = 78$ nm; (b) $B = 636$ G, $a = 35$ nm. Note the difference in time scales. Solid lines are fits with two-body and one-body processes giving initial time constants of 500 ms in (a) and 20 ms in (b).

whereas Fig. 4(b) corresponds to $B = 636$ G, $a = 35$ nm, and $E_b/k_B = 60$ μK. The initial fraction of unpaired atoms (not engaged in a molecule) is 17% in (a) and 25% in (b). For a change in a by the modest factor of 2.2, the two samples exhibit strikingly different lifetimes. Decays are well fitted by two-body and one-body loss processes with initial lifetimes of $\simeq 500$ ms in (a) (689 G) and 20 ms in (b) whereas the one-body decay exceeds 4 s.

A strong decrease in the lifetime of molecules with decreasing a is expected assuming collisional relaxation to deep bound states [25]. However, the observed factor of 25 in lifetimes is surprisingly large. At 689 G, we can evaluate the two-body loss rate to be $G = 2.4^{+3.2}_{-1.6} \times 10^{-13}$ cm^3 s^{-1}. At 636 G, we have initially $N_3 \simeq 0.3 N_1$. These fast losses indicate that very likely some evaporation is involved in the limited lifetime and that the system has not reached thermal equilibrium. Therefore, the two-body loss rate cannot be safely estimated.

Let us now evaluate the phase-space density of the trapped molecules. In Fig. 2, at 689 G, there are 1.8×10^4 molecules, confined with 3.3×10^4 atoms. The measured atom temperature is $T_{at} = 6.7$ μK. Assuming thermal equilibrium between atoms and molecules, $T_{at} = T_m$ for magnetic fields above $\simeq 650$ G, we obtain the peak density of molecules $n_m \simeq 4 \times 10^{13}$ cm^{-3}. Then the critical temperature for molecule condensation is reduced due to the interactions and is 3.5 μK giving $T/T_C \simeq 2$. Since $n_m a_m^3 \simeq 5 \times 10^{-3} \ll 1$ with $a_m = 0.6a = 47$ nm [25], the molecular gas is in the dilute regime and the mean distance between molecules is larger than the typical size of a molecule a. In fact, for all data between 675 and 750 G, the phase-space density of the molecules is not far from the condensation point. Since, the lifetime of the molecules, 500 ms, is long when compared to the molecule-molecule elastic collision time $1/(n_m 8\pi a_m^2 v_m) \simeq 3$ μs (for $a_m = 47$ nm), it should be possible to evaporate the molecules further to reach the Bose-Einstein condensation threshold.

In summary, we have produced long-lived and trapped Li_2 dimers. The atom-molecule conversion efficiency can approach 1 when the Fermi quantum degeneracy is strong. The lifetime of the trapped molecules strongly depends on the scattering length. Current research concentrates on evaporation towards molecular BEC. Prospects for producing superfluid Fermi mixtures and for investigating the transition between molecular condensates and superfluid Fermi gases are promising [11,12,25,26]. Long lifetimes in ^{6}Li [27,28] and ^{40}K [29] dimers have also been recently observed.

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