A molecular Bose-Einstein condensate emerges from a Fermi sea

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(Dated: November 3, 2003)

The realization of fermionic superfluidity in a dilute gas of atoms, analogous to superconductivity in metals, is a long-standing goal of ultracold gas research. Beyond being a new example of this fascinating quantum phenomenon, fermionic superfluidity in an atomic gas holds the promise of adjustable interactions and the ability to tune continuously from BCS-type superfluidity to Bose-Einstein condensation (BEC). This crossover between BCS superfluidity of correlated atom pairs in momentum space and BEC of local pairs has long been of theoretical interest, motivated in part by the discovery of high $T_c$ superconductors.\textsuperscript{1, 2, 3} In atomic Fermi gas experiments superfluidity has not yet been demonstrated; however recent experiments have made remarkable progress toward this goal. Starting from an ultracold Fermi gas experimenters have used Feshbach resonances to reversibly create molecules, i.e. composite bosons consisting of local fermion pairs.\textsuperscript{4, 5, 6, 7} Furthermore, the experiments have shown that the resulting diatomic molecules can have surprisingly long lifetimes.\textsuperscript{8, 9, 10} Here we report the conversion of a Fermi sea of atoms into a molecular BEC. In addition to being the first molecular condensate in thermal equilibrium, this BEC represents one extreme of the predicted BCS-BEC crossover.

The basic idea behind this experiment is to start with a Fermi gas that has been evaporatively cooled to a high degree of quantum degeneracy and adiabatically create molecules with a magnetic-field sweep across the Feshbach resonance. If the molecule creation conserves entropy and the initial atom gas is at sufficiently low temperature $T$ compared to the Fermi temperature $T_F$, then the result should be a molecular sample with a significant condensate fraction.\textsuperscript{11, 12} With a relatively slow sweep of an applied magnetic field that converts most of the fermionic atoms into bosonic molecules and an initial atomic gas below $T/T_F = 0.17$, we observe a molecular condensate in time-of-flight absorption images taken immediately following the magnetic-field sweep. Strikingly, the molecular condensate is not formed by any active cooling of the molecules, but rather merely by traversing the predicted BCS-BEC crossover regime.

Our experimental set-up and procedure used to cool a gas of fermionic $^{40}$K atoms to quantum degenerate temperatures is detailed in previous work.\textsuperscript{13, 14, 15} In brief, after laser cooling and trapping we evaporatively cool the atoms in a magnetic trap. In order to realize $s$-wave collisions in the ultracold Fermi gas we use a mixture of atoms in two different spin states. For the final stage of evaporative cooling the atoms are loaded into an optical dipole trap formed by a single far red-detuned laser beam. The laser wavelength is $\lambda = 1064$ nm and the beam is focused to a waist of 15.5 $\mu$m. By lowering the depth of the optical trap we evaporate the atomic gas to temperatures far below the Fermi temperature $T_F = (6N\nu_r^2\nu_z)^{1/3}h/k_B$. Here $N$ is the particle number in each spin state, $\nu_r$ and $\nu_z$ are the radial and axial trap frequencies, $h$ is Planck’s constant, and $k_B$ is Boltzmann’s constant. For final radial trap frequencies between $\nu_r = 430$ Hz and 250 Hz and a fixed trap aspect ratio $\nu_r/\nu_z = 79 \pm 15$, we achieve temperatures between $0.36 T_F$ and $0.04 T_F$. All temperatures of the Fermi gas given in this work are determined through surface fits to time-of-flight absorption images.\textsuperscript{16}

For this work we use a Feshbach resonance, which occurs when the energy of a quasibound molecular state becomes equal to the energy of two free atoms.\textsuperscript{17} The magnetic-field dependence of the resonance allows for precise tuning of the atom-atom interaction strength in an ultracold gas.\textsuperscript{18, 19} Moreover, time-dependent magnetic fields can be used to reversibly convert atom pairs into extremely weakly bound molecules.\textsuperscript{20, 21, 22, 23, 24} The particular resonance used here is located at a magnetic field $B_0 = 202.1 \pm 0.1$ G and has a width of $w = 7.8 \pm 0.6$ G.\textsuperscript{14, 25} The resonance affects collisions between atoms in the two lowest energy spin states $|f = 9/2, m_f = -7/2\rangle$ and $|f = 9/2, m_f = -9/2\rangle$, where $f$ denotes the total atomic angular momentum and $m_f$ the magnetic quantum number.

To create bosonic molecules from the fermionic atoms, we first prepare an equal mixture of atoms in the $m_f = -9/2$ and $m_f = 7/2$ spin states at temperatures below quantum degeneracy. Then we apply a time-dependent sweep of the magnetic field starting above the Feshbach resonance value, where the atom interactions are effectively attractive, and ending below the resonance, where the atom interactions are effectively repulsive. In contrast to our previous work\textsuperscript{10} the magnetic-field sweep is not only adiabatic with respect to the molecule creation rate, but also slow with respect to the collision rate and the radial trap frequency.\textsuperscript{12} The magnetic field is typically ramped in 7 ms from $B = 202.78$ G to either $B = 201.54$ G or $B = 201.67$ G. With this magnetic-field sweep across the Feshbach resonance we convert between 78% and 88% of the atoms into molecules. To a very
A critical element of this experiment is that the lifetime of these weakly bound molecules can be much longer than the typical collision time in the gas and longer than the radial trapping period.\cite{11-14} In previous work we found that the $^{40}$K$_2$ molecule lifetime increases dramatically near the Feshbach resonance and reaches $\sim$100 ms at a magnetic field 0.43 G below the Feshbach resonance for a peak density of $n_{pk} = 1.5 \times 10^{13}$ cm$^{-3}$.\cite{11-14} It is predicted that this increased molecule lifetime only occurs for dimers of fermionic atom pairs.\cite{31} The relatively long molecule lifetime near the Feshbach resonance allows the atom/molecule mixture to achieve thermal equilibrium during the magnetic-field sweep. Note however that the large aspect ratio of the optical trap gives rise to a strongly anisotropic system. Thus for the relatively short timescale of the experiments reported here we may attain only local equilibrium in the axial direction.\cite{32}

To study the resulting atom-molecule gas mixture after the magnetic-field sweep, we measure the momentum distribution of both the molecules and the residual atoms using time-of-flight absorption imaging. After typically 10 to 20 ms of expansion we apply a radio frequency (rf) pulse that dissociates the molecules into free atoms in the $m_f = 5/2$ and $m_f = 9/2$ spin states.\cite{10} Immediately after this rf dissociation pulse we take a spin-selective absorption image. The rf pulse has a duration of 140 $\mu$s and is detuned 50 kHz beyond the molecule dissociation threshold where it does not affect the residual unpaired atoms in the $m_f = 7/2$ state. We selectively detect the expanded molecule cloud by imaging atoms transferred by the rf dissociation pulse into the previously unoccupied $m_f = 5/2$ state. Alternatively we can image only the expanded atom cloud by detecting atoms in the $m_f = 7/2$ spin state.

Close to the Feshbach resonance, the atoms and molecules are strongly interacting with effectively repulsive interactions. The scattering length for atom-molecule and molecule-molecule collisions close to the Feshbach resonance has recently been calculated by Petrov et al.\cite{29} to be 1.2 $a$ and 0.6 $a$ respectively, where $a$ is the atom-atom scattering length.\cite{31} During the initial stage of expansion the positive interaction energy is converted into additional kinetic energy of the expanding cloud. Therefore the measured momentum distribution is very different from the original momentum distribution of the trapped cloud. In order to reduce the effect of these interactions on the molecule time-of-flight images we use the magnetic-field Feshbach resonance to control the interparticle interaction strength during expansion. We can significantly reduce the interaction energy momentum kick by rapidly changing the magnetic field before we switch off the optical trap for expansion. The field is lowered typically by 4 G in 10 $\mu$s. At this magnetic field further away from the resonance the atom-atom scattering length $a$ is reduced to $\sim$500 $a_0$. We find that this magnetic-field jump results in a loss of typically 50% of the molecules, which we attribute to the reduced molecule lifetime away from the Feshbach resonance.

A good approximation these molecules have twice the polarizability of the atoms\cite{29} and therefore are confined in the optical dipole trap with the same trapping frequency and twice the trap depth of the atoms. The molecules, which are all in the same internal quantum state, are highly vibrationally excited, very large in spatial extent, and extremely weakly bound. For a magnetic field 0.43 G below the Feshbach resonance ($B=201.67$ G) the binding energy determined through a full coupled channels calculation is 8 kHz\cite{30} and the molecule size is $\sim$1650 $a_0$, where $a_0$ is the Bohr radius. Here we estimate the molecule size as $a/2$, where $a$ is the atom-atom scattering length given by $a = 174a_0[1 + w/(B_0 - B)]$. At this magnetic field the molecule size is one order of magnitude smaller than the calculated intermolecular distance.
Below an initial temperature of 0.17 $T_F$ we observe the sudden onset of a pronounced bimodal momentum distribution for the molecules. Figure 1 shows such a bimodal distribution for an experiment starting with an initial temperature of 0.06 $T_F$; for comparison we also show the resulting molecule momentum distribution for an experiment starting at 0.19 $T_F$. The bimodal momentum distribution is a striking indication that the cloud of weakly bound molecules has undergone a phase transition to a Bose-Einstein condensate.\textsuperscript{33--35}

In order to obtain thermodynamic information about the molecule cloud we fit the momentum distribution with a two-component fit. The fit function is the sum of an inverted parabola describing the Thomas-Fermi momentum distribution of a bosonic condensate and a Gaussian momentum distribution describing the noncondensed component of the molecule cloud. In Fig. 2 the measured condensate fraction is plotted as a function of the fitted temperature of the thermal component in units of the critical temperature for an ideal Bose gas $T_c = 0.94(N v^2 \nu_c)^{1/3} h/k_B$. Here $N$ is the total number of molecules measured without changing the magnetic-field for the expansion. Note that this measurement may underestimate the original condensate fraction due to loss of molecules during expansion. From Fig. 2 we determine an actual critical temperature for the strongly interacting molecules and for our trap geometry of $0.8 \pm 0.1 T_c$. Such a decrease of the critical temperature relative to the ideal gas prediction is expected for a strongly interacting gas.\textsuperscript{36}

We find that the creation of a Bose-Einstein condensate of molecules requires that the Feshbach resonance be traversed sufficiently slowly. This is illustrated in Fig. 3, where the measured condensate fraction is plotted versus the ramp time across the Feshbach resonance starting with a Fermi gas at a temperature 0.06 $T_F$. Our fastest sweeps result in a much smaller condensate fraction while the largest condensate fraction appears for a B-field sweep of 3 to 10 ms. For even slower magnetic field sweeps we find that the condensate fraction slowly decreases. We attribute this effect to a finite lifetime of the condensate. Note that the timescale of the experiment is short compared to the axial trap frequency. Therefore the condensate may not have global phase coherence in the axial direction.\textsuperscript{32} The inset of Fig. 3 shows a plot of the lifetime of the condensate. The observed reduction in condensate fraction is accompanied by heating of the molecule gas, presumably due to collisional decay of the molecules into more tightly bound states.

Rapidly changing the interaction strength for time-of-flight expansion of the condensate allows us to measure the interaction energy in the molecular sample. Figure 4 shows a plot of the expansion energy of the molecule BEC for various interaction strengths during time-of-flight expansion. Here the condensate is created at a fixed interaction strength, and thus the initial peak density $n_{pk}$ is constant. The data show that the expansion energy is proportional to the atom-atom scattering length. The linear dependence suggests that the molecule-molecule scattering length is proportional to the atom-atom scattering length as predicted in Ref. 31. In addition we find that the expansion energy extrapolates to near zero energy for $a=0$. This is consistent with a Bose-Einstein condensate of molecules. Assuming the molecule-molecule interaction strength calculated in Ref. 31 this measurement allows us to determine the peak density of the

![FIG. 2: Molecular condensate fraction $N_0/N$ versus the scaled temperature $T/T_c$. The temperature of the molecules is varied by changing the initial temperature of the fermionic atoms prior to the formation of the molecules. All other parameters are similar to the ones described in the caption of Fig. 1. We observe the onset for Bose-Einstein condensation at a temperature of $\sim 0.8 T_c$.](image1)

![FIG. 3: Dependence of condensate formation on magnetic-field sweep rate and measurement of condensate lifetime. We plot the fraction of condensed molecules versus the time in which the magnetic field is ramped across the Feshbach resonance from 202.78 G to 201.54 G. The condensate fraction is measured after an additional waiting time of 1 ms. The initial atom gas temperature is 0.06 $T_F$, the total molecule number is 150,000, and the final radial trap frequency is 260 Hz. For the full range of ramp times the number of molecules created remains constant. In the inset the condensate fraction is plotted versus the wait time after a 10 ms magnetic-field ramp. The molecule number is not significantly reduced on this timescale, and the lifetime of the condensate is instead determined by a heating rate, which we measure to be $3 \pm 1$ nK/ms. This heating rate is presumably due to density dependent inelastic loss processes.](image2)
strongly interacting condensate as \( n_{pk} = 7 \times 10^{12} \text{ cm}^{-3} \).

A fundamental aspect of the experiment is that we start with a quantum degenerate Fermi gas of atoms. The Bose-Einstein condensate, which is observed immediately after the magnetic field is ramped across the Feshbach resonance, therefore requires a drastic change of the quantum statistical thermodynamics of the gas. This change is not due to evaporative cooling and the total number of atoms (adding both free atoms and those bound in molecular dimers) is conserved by the field sweep. In Figure 5 we show the dependence of the condensate fraction on the initial temperature of the Fermi gas. We find that a BEC is formed when the initial temperature is below 0.17 \( T_F \). If we assume that entropy is conserved in the sweep across the Feshbach resonance, then creating the molecular Bose-Einstein condensate depends on starting with a Fermi gas at sufficiently low \( T/T_F \) to give low initial entropy.\(^\text{15} \) At the onset of BEC our temperature measurement indicates a modest 40\% increase in the total entropy after the magnetic field sweep, estimated from an ideal gas model. The inset in Fig. 5 compares the absolute temperature of atoms and molecules after the magnetic field sweep. For the molecules the temperature is determined by a fit to the non-condensate fraction. We find that atoms and molecules are well thermalized. Note that the atoms and molecules are not in full chemical equilibrium.\(^\text{12} \) Even though the final binding energy of the molecules is significantly larger than kinetic energy in the gas, we only observe conversion efficiencies of up to 88\%. In order to study the reversibility of the ramp across the Feshbach resonance we have ramped the magnetic field back to the attractive side of the resonance after creating a molecular condensate and then measured the temperature of the resulting Fermi gas. We find that the gas is heated by 27 ± 7 nK in this double ramp, independent of the initial temperature.

In conclusion, we have created a Bose-Einstein condensate of weakly bound molecules starting with a gas of ultracold fermionic atoms. The molecular BEC has been detected through a bimodal momentum distribution, and effects of the strong interparticle interaction have been investigated. The molecular BEC reported here, which appears on the repulsive side of the Feshbach resonance, is related in a continuous way to BCS-type fermionic superfluidity on the attractive side of the resonance. Our experiment corresponds to the BEC limit, in which superfluidity occurs due to BEC of essentially local pairs whose binding energy, \( \sim \hbar^2/m a^2 \), is much larger than the Fermi energy. Here \( m \) is the atomic mass and \( 2\pi\hbar \) is Planck’s constant. The dimensionless parameter \( 1/k_F a \), which drives the crossover from a BCS-superfluid to a molecular BEC\(^\text{3,4} \) is thus positive and large compared to one. In this regime, the formation of molecules is clearly separated from their condensation. In contrast, near the Feshbach resonance where \( 1/k_F a \) goes through zero, the system may be described neither by a BEC of molecular dimers nor a BCS-state of correlated pairs in momentum space, a situation which has been termed “resonance superfluidity”.\(^\text{6} \) Indeed our experiment passes through this largely unexplored crossover regime and with initial temperatures below 0.1 \( T_F \) the Fermi gas is well below the predicted critical temperature in the crossover regime of 0.22 \( T_F \).\(^\text{9} \) In future work it will be very exciting to investigate this system on the attractive side of the resonance.
and look for evidence of fermionic superfluidity.

We thank L. D. Carr, E. A. Cornell, C. E. Wieman, W. Zwerger, and I. Bloch for useful discussion and J. Smith for experimental assistance. This work was supported by NSF and NIST, and C. A. R. acknowledges support from the Hertz Foundation.

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