Creation of ultracold molecules from a Fermi gas of atoms

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Since the realization of Bose-Einstein condensates (BEC) in atomic gases an experimental challenge has been the production of molecular gases in the quantum regime. A promising approach is to create the molecular gas directly from an ultracold atomic gas; for example, atoms in a BEC have been coupled to electronic ground-state molecules through photoassociation \[ \text{as well as through a} \]

magnetic-field Feshbach resonance \[ \text{The availability of atomic Fermi gases provides the exciting prospect of coupling fermionic atoms to bosonic molecules, and thus altering the quantum statistics of the system. This Fermi-Bose coupling is closely related to the pairing mechanism for a novel fermionic superfluid proposed to occur near a Feshbach resonance \[ \text{Here we report the creation and quantitative characterization of exotic, ultracold }^{40}\text{K}_2 \text{molecules. Starting with a quantum degenerate Fermi gas of atoms at } T < 150 \text{ nanoKelvin we scan over a Feshbach resonance to adiabatically create over a quarter million trapped molecules, which we can convert back to atoms by reversing the scan. The small binding energy of the molecules is controlled by detuning from the Feshbach resonance and can be varied over a wide range. We directly detect these weakly bound molecules through rf photodissociation spectra that probe the molecular wavefunction and yield binding energies that are consistent with theory.} \]

Scattering resonances known as Feshbach resonances occur when the collision energy of two free atoms coincides with that of a quasi-bound molecular state \[ \text{By varying the strength of an external magnetic field experimenters can tune the relative atom-molecule energy through the Zeeman effect. This enables control over the strength of cold atom interactions, characterized by the s-wave scattering length } a, \text{ as well as whether they are effectively repulsive } (a > 0) \text{ or attractive } (a < 0). \text{ These resonances have been used to tune the interaction strength between atoms for both Bose and Fermi gases} \[ \text{Another possible use of a Feshbach resonance is to controllably convert atoms into molecules. The energy of the molecular state associated with the Feshbach resonance coincides with the free atom threshold at the resonance peak, and the binding energy of the molecule varies smoothly with magnetic field on the repulsive side of the resonance } (a > 0). \text{ Thus, one would expect that atoms could be coupled to molecules by ramping the detuning from the resonance using a time-dependent magnetic field} \[ \text{For example, ramping the magnetic field through a Feshbach resonance resulted in large losses in a }^{23}\text{Na BEC} \]. \text{Further, coherent oscillations between atoms and molecules in a BEC were observed following magnetic field pulses on the repulsive side of a }^{85}\text{Rb Feshbach resonance} \]. \text{Here we report the efficient creation of bosonic molecules from fermionic }^{40}\text{K atoms using a magnetic field ramp across a Feshbach resonance. We present clear evidence of diatomic molecule formation through direct, spectroscopic detection of these molecules.} \[ \text{The experiments reported here employ previously developed techniques for cooling and spin state manipulation of }^{40}\text{K} \]. \text{Because of the quantum statistics of fermions a mixture of two components, for example atoms in different internal spin states, is required to have s-wave interactions in the ultracold gas. With a total atomic spin } J = 9/2 \text{ in its lowest hyperfine ground state, }^{40}\text{K has ten available Zeeman spin-states } | f, m_f \rangle, \text{ where } m_f \text{ is the spin projection quantum number. Mixtures of atoms in two of these spin states are used in evaporative cooling of the gas, first in a magnetic trap and then in a far-off resonance optical dipole trap. The optical trapping potential has the distinct ability to trap atoms in any spin state as well as any molecules created from these atoms. For these experiments the optical trap is characterized by radial frequencies ranging from } \nu_r = 215 \text{ to 276 Hz, with the trap aspect ratio, } \nu_z/\nu_r, \text{ fixed at } 70 \pm 20. \text{ The temperature } T \text{ of these two-component gases, measured in units of the Fermi temperature } T_F, \text{ ranges from } T/T_F = 0.13 \text{ to 0.33. This degree of quantum degeneracy is near the lowest ever demonstrated in a Fermi gas of atoms} \]. \text{Experiments are initiated by preparing atoms in a nearly equal, incoherent mixture of the } |9/2, -5/2 \rangle \text{ and } |9/2, -9/2 \rangle \text{ spin states. With these states we access a previously reported Feshbach resonance located at a magnetic field of } 224.21 \pm 0.05 \text{ G (Fig. 1d)} \]. \text{A time-dependent current through an auxiliary coil provides magnetic field ramps near the resonance. Starting from a magnetic field of } 227.81 \text{ G, the field is ramped at a rate of } (40 \text{ } \mu\text{s}/\text{G})^{-1} \text{ across the resonance to various final values. The number of atoms remaining following the ramp is determined from an absorption image of the cloud (at } \sim 4 \text{ G) after expansion from the optical trap. Since the light used for these images is resonant with the atomic transition, but not with any molecular transitions, we selectively detect only the atoms. In Fig. 1 we present the observed atom number as a function of} \]
the final magnetic field value of the ramp. We find that the atoms disappear abruptly at the Feshbach resonance peak.

We also investigate the atom loss as a function of the rate of the magnetic field sweep. Fig. 2 illustrates the result of linear magnetic field ramps across the Feshbach resonance from 228.25 G to 216.15 G. For our fastest sweeps there is no observable effect upon the atoms, while significant atom number loss is observed for slower sweeps. For our slowest sweeps we find that the number of atoms lost saturates at 50%. The atoms vanish at least two orders of magnitude more quickly than expected from previously measured inelastic collision rates at a resonance [13]. When we reverse the process by applying an additional magnetic field ramp across the Feshbach resonance in the opposite direction, we observe the return of nearly all the “lost” atoms (Fig. 2). This is consistent with the lost atom number corresponding to trapped molecules.

Surprisingly, the number of molecules produced is very large despite the fact that the Fermi gas is not described by a single wavefunction as is a BEC. In fact the number of molecules is sufficiently large that the temperature of our initial atomic gas is below the molecular Bose-Einstein condensation temperature in the optical trap. Further, we can measure the lifetime $\tau$ of the molecules by varying the time before conversion back to atoms; we find that $\tau \sim 1 \text{ ms}$.

Using radio frequency spectroscopy we directly probe these molecules. First, we create the molecules with a $\left(40 \text{ } \mu\text{s}/\text{G}\right)^{-1}$ magnetic field ramp. This ramp starts at 227.81 G and ends at various final magnetic field values $B_{\text{hold}}$ below the resonance. At $B_{\text{hold}}$ a 13 $\mu$s rf pulse is applied to the cloud; the rf frequency is chosen so that the photon energy is near the energy splitting between the $|9/2, -5/2\rangle$ and $|9/2, -7/2\rangle$ atom states. The resulting population in the $|9/2, -7/2\rangle$ state, which is initially unoccupied, is then probed selectively either by separating the spin states spatially using a strong magnetic field gradient during free expansion (Stern-Gerlach imaging), or by leaving the magnetic field high (215 G) and taking advantage of nonlinear Zeeman shifts.

Fig. 3 shows a sample rf spectrum at $B_{\text{hold}} = 222.49$ G; the resulting number of atoms in the $|9/2, -7/2\rangle$ state is plotted as a function of the frequency of the rf pulse. We observe two distinct features in the spectrum. The sharp, symmetric peak is very near the expected $|9/2, -5/2\rangle$ to $|9/2, -7/2\rangle$ transition frequency for free atoms $\nu_{\text{atom}}$. With the Stern-Gerlach imaging we see that the total number of $|9/2, -5/2\rangle$ and $|9/2, -7/2\rangle$ atoms is constant, consistent with transfer between these
two atom states. The width of this line is defined by the fourier width of the applied rf pulse. Nearby is a broader, asymmetric peak shifted lower in frequency. Here we find that after the rf pulse the total number of observed atoms \((|9/2, -7/2\rangle + |9/2, -9/2\rangle\) actually increases (Fig. 3b). Also, the resulting \(|9/2, -7/2\rangle\) gas in this region has a significantly increased kinetic energy, which grows linearly for larger frequency shifts from the atom peak (Fig. 3b).

The asymmetric peak can be interpreted as the dissociation of molecules into \(|9/2, -7/2\rangle\) and \(|9/2, -9/2\rangle\) atoms. Since the applied rf pulse stimulates a transition to a lower energy Zeeman state, we expect \(h\nu_f = h\nu_0 - E_{\text{binding}} - \Delta E\), where \(h\) is Planck’s constant, \(E_{\text{binding}}\) is the binding energy of the molecule, and we have ignored mean-field interaction energy shifts. The remaining energy, \(\Delta E\), must by imparted to the dissociating atom pair as kinetic energy. Further, the observed lineshape of the asymmetric peak should depend on the Franck-Condon factor, which gives the overlap of the molecular wavefunction with the atomic wavefunction.

We have calculated this multichannel Franck-Condon overlap as a function of energy. The resulting transition rate, convolved with the frequency width of the applied rf and scaled vertically, is shown as the solid line in Fig. 3a. The agreement between theory and experiment for the dissociation spectrum is quite good. This well-resolved spectrum provides much information about the molecular wavefunction. For example, the mean interatomic separation of the molecules at this magnetic field is extremely large, \(\sim 170a_0\), where \(a_0\) is the Bohr radius.

In Fig. 4 we plot the magnetic field dependence of the frequency shift \(\Delta\nu\) between the atom line and the threshold of the molecular spectrum, which should correspond to the molecular binding energy. While this could in principle be obtained directly from the transfer spectrum (Fig. 4b), the appearance of the threshold in the energy of the \(|9/2, -7/2\rangle\) cloud is more clear (Fig. 4c). We compare the position of this energy threshold to the expected atom-atom transition frequency \(\nu_{\text{atom}}\) based upon a calibration of the magnetic field strength. The data are consistent with a theoretical calculation of the binding energy (solid line) based upon a full coupled channels calculation with no free parameters.

This agreement with theory leaves no doubt that we are creating large numbers of weakly bound molecules. These highly vibrationally excited molecules could be used to study ultracold molecule-molecule or molecule-atom collisions. Further, the explicit coupling of a quantum degenerate gas of Fermi atoms to bosonic molecules could possibly be developed as a method to facilitate creation of a predicted exotic fermionic superfluid. In addition our molecule detection technique could be extended to measure the gap energy in this superfluid phase.
FIG. 5: Binding energy of the molecules. The frequency shift ($\Delta \nu$) from the expected $|9/2, -5/2 \rangle \rightarrow |9/2, -7/2 \rangle$ transition is plotted versus magnetic field for the $|9/2, -7/2 \rangle$ atoms (squares) and the molecules (circles). The typical atom cloud before molecule creation is characterized by $T/T_F = 0.14$ and $N = 7 \times 10^5$. The dashed line indicates the Feshbach resonance position. The solid line is a calculation of the binding energy of the molecules as a function of detuning from the resonance. The small shift in the atom transition frequency is due to the atom-atom interaction energy.

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