Sensitive Detection of Cold Cesium Molecules Formed on Feshbach Resonances

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We observe the dynamic formation of quasibound Cs_2 molecules near Feshbach resonances in a cold sample of atomic cesium. Using an external probe beam, more than 15 weakly coupled molecular states are detected with high sensitivity, whose collisional formation cross sections are as small as $\sigma = 2 \times 10^{-16}$ cm^2. By modeling the molecule formation and dissociation processes with rate equations, we conclude that at an atomic density of 10^{13} cm^{-3} and temperature of 5 μK, more than $5(1) \times 10^5$ Cs$_2$ molecules in a single rovibrational state coexist with 10^8 Cs atoms in our trap.

The newly emerging field of cold molecules provides intriguing possibilities in ultracold gas studies. In the realm of "superchemistry," phase-coherent chemical reactions between particles in well-defined internal and external states are dominated by quantum statistics and many-body effects [1]. The coherent nature of the atom-molecule coupling has been recently observed in Bose-Einstein condensates using a Feshbach resonance [2]. Quantum manipulation of molecular states has also been proposed for quantum computation [3]. Finally, in precision tests of time-reversal symmetry, polar molecule experiments promise a sensitivity several orders of magnitude higher than atoms [4].

Recently, macroscopic numbers of cold molecules have been produced with several methods. Starting from a cold atomic gas in a magneto-optical trap (MOT), 2×10^5 molecules have been created in a few vibrational states and magnetically trapped [5]. Two-photon photoassociation from an atomic condensate allows coupling to a single high-lying molecular state, although inelastic losses limit the molecule number [6]. Alternative routes to cold molecules by slowing down a supersonic molecular jet [7], or by buffer gas loading of a magnetic trap [8], can collect up to 10^9 molecules, but the resulting temperatures are higher than in the approaches using cold atoms.

A molecular population can also be created in a cold atomic sample near a Feshbach resonance, where the atom-atom scattering continuum $|k\rangle$ couples directly to a molecular state $|m\rangle$ supported by a closed scattering channel with a higher asymptotic energy [9], as shown in Fig. 1. In this case the molecular state becomes quasibound, acquiring a width determined by the coherent coupling strength and the scattering energy, even in the absence of inelastic processes [9,10]. The resulting dynamic equilibrium between atomic and molecular populations resembles that which occurs between ion-electron pairs and neutral atoms in the stellar plasma, as described by the Saha-Boltzmann equation [11]. It is shown below that for harmonically trapped atoms with peak density n_0 and phase space density $\phi = n_0 \lambda_{\mathrm{dB}}^3$,

where λ_{dB} is the thermal de Broglie wavelength, a peak molecular density as large as $m_0 = n_0 \phi$ can be reached near a Feshbach resonance.

In this work, we observe the dynamic formation of quasibound Cs_2 molecules near narrow Feshbach resonances. The observed linewidths of these resonances as small as $\delta\omega/2\pi \sim 5$ kHz [12], and the negligible resonant loss, suggest that the associated molecular states are only weakly and coherently coupled to the scattering continuum with a lifetime $\delta t \gtrsim 30$ μs. A large molecular population can therefore build up in our thermal sample. Using a rate equation model, and an upper bound on the inelastic collision rate deduced from the unitarity limit, we determine a lower bound on the number of quasibound molecules coexisting with the atoms, which agrees with the prediction from the Saha equation.

To monitor the formation of molecules, we tune the atomic collisions to Feshbach resonance using an external magnetic field [9] and employ a probe beam to excite and dissociate the quasibound molecules, as shown in Fig. 1.
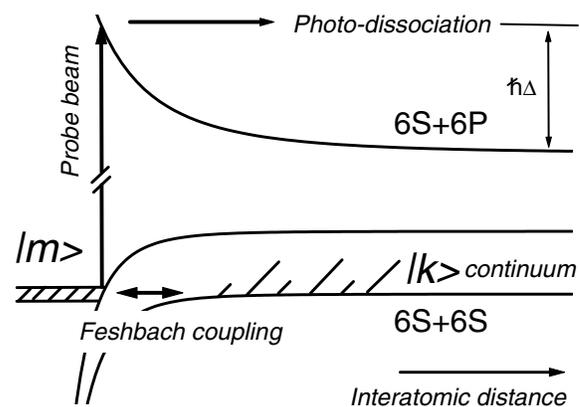

FIG. 1. Illustration of our detection method. In the vicinity of a Feshbach resonance the ground molecular state $|m\rangle$ is coupled to the scattering continuum $|k\rangle$. A probe beam tuned to the blue of the free-atom transition excites only the molecules, which then dissociate and are lost from the trap due to the energy imparted by the strongly repulsive excited-state potential.

This method is similar to the technique first introduced by Heinzen and coworkers, in which a laser detuned from the free-atom resonance is used as a sensitive probe of resonant changes in the short-range two-atom correlation function [13]. However, instead of using a red-detuned laser to excite the atom pairs into excited molecular states, we tune our probe laser far above the free-atom resonance where the interatomic potentials are purely repulsive due to the dipole-dipole interaction. Atom pairs are preferentially excited at a particular interatomic distance where the electronic transition is shifted into resonance by this interaction, and the interatomic force at these distances is sufficiently strong that excited atom pairs are expelled from our trap. Furthermore, we can selectively excite the molecules, while leaving the free atoms unaffected, by choosing a sufficiently large detuning Δ so that the excitation occurs at a very short range where free-atom pairs are extremely unlikely to be found. The laser-induced depletion of the molecules formed on a Feshbach resonance provides an inelastic decay channel for the trapped atoms, and the resultant atom loss constitutes a highly sensitive and background-free probe of the Feshbach resonance and of the molecular population. This method has the advantage that it requires no prior knowledge of the photoassociation spectrum, and it may be of interest as a general technique for heteronuclear collisions between different atomic species, where the interatomic potentials are not known in detail beforehand.

Compared to lighter alkalis, cesium atoms display unusually rich cold-collision phenomena, due primarily to the large dipolar interaction which allows coupling to molecular states with high rotational angular momentum [14]. It is precisely this interaction that produces the extremely narrow Feshbach resonances observed in this work which result from coupling of incoming s and p partial waves to rovibrational molecular states with rotational angular momentum up to $4\hbar$ [15]. We focus here on the two lowest-energy collision channels, where either both colliding atoms are in the ground state $|F = 3, m_F = 3\rangle$, or one atom is in $|F = 3, m_F = 3\rangle$, and the other in $|F = 3, m_F = 2\rangle$, and we denote these two channels below by $|3, 3\rangle + |3, 3\rangle$ and $|3, 3\rangle + |3, 2\rangle$, respectively. The negligible inelastic cross section in these two channels [15] are necessary to allow a large equilibrium molecular population to build up in our sample.

Our experimental setup has been described previously in Ref. [16]. 3×10^8 atoms are loaded into a far-detuned dipole trap formed by a Nd:YAG laser at 1064 nm using two pulses of Raman-sideband cooling [16,17]. We then further optically pump so that up to 98% of the remaining atoms are in $|3, 3\rangle$, and by adjusting the optical pumping frequency we prepare mixed samples as desired with up to 10% in $|3, 2\rangle$. For all of the experiments described here, less than 1% (0.25%) of the atoms are found to occupy $|3, 1\rangle$ (all other states), as determined by microwave spec-

troscopy [18]. Within 15 ms the atoms thermalize to a temperature between 3 and 6 μK and a density near 10^{13} cm^{-3} by means of elastic collisions; the atom number is measured by fluorescence detection and the density is then calculated from the measured temperatures and trap vibration frequencies [16]. We adiabatically ramp the magnetic field from 0.1 to 2 G in 1 ms, and then to any desired field value up to 230 G in another 1 ms. No depolarization of the sample is observed during this procedure. A titanium-sapphire laser provides the far-detuned probe beam with an e^{-2} beam waist of 2.0 mm (0.6 mm) in the vertical (horizontal) direction, which uniformly illuminates the sample whose vertical (horizontal) size is 580 μm (60 μm) [12]. The typical mean intensity and wavelength, optimized for radiative detection sensitivity, are 20 W/cm^2 and 847 nm, respectively. Loss coefficients are extracted from the evolution of the atomic density, assuming a Gaussian distribution with constant temperature, according to $\dot{n} = -n/T - Gn^2 - (4/3)^{3/2} K n^3$, where n is the mean atomic density, $T = 3$ s is the measured one-body lifetime, and G (K) is the two-(three-)body decay coefficient.

Figure 2 shows the radiative loss rate between 0 and 230 G, measured with a resolution of 100 mG. All seven strong resonances shown are due to $|3, 3\rangle + |3, 3\rangle$ collisions. The lower curve in the inset shows a higher-resolution scan performed on a sample with a larger impurity in $|3, 2\rangle$. Seven additional smaller resonances are evident, marked by asterisks, which originate from $|3, 3\rangle + |3, 2\rangle$ collisions. The upper curve in the inset is the corresponding measurement of elastic-collision-induced evaporative loss performed under similar conditions [18]: a reduction in elastic collision rate due to a
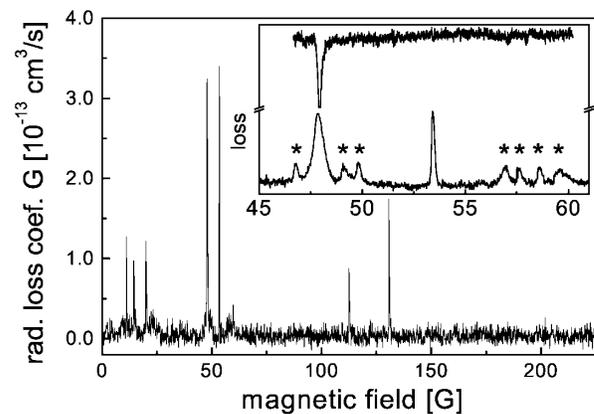

FIG. 2. Radiative loss coefficient as a function of magnetic field for a mean density $n = 1.5 \times 10^{13}$ cm^{-3}, 95% population in $|F = 3, m_F = 3\rangle$, and a temperature of 5.5 μK. The wavelength and the mean intensity of the probe beam are 846.52 nm and 33 W/cm^2, respectively. In the inset, the radiative loss spectrum (lower curve) shows higher sensitivity than the collision rate measurement (upper curve). The asterisks indicate $|3, 3\rangle + |3, 2\rangle$ collision resonances.

Feshbach resonance increases the thermalization time of the sample and results in a reduced evaporation loss of atoms from a trap of finite depth. Of all the resonant features observed here, only the strongest one at 48.0 G was also detected as a change in the elastic collision rate. This occurs because the variation of the elastic cross section near a narrow resonance is averaged away by the thermal distribution of collision energies. Finally, in the absence of the probe beam, we observe no resonant three-body recombination loss near these narrow resonances within our sensitivity of $K = 3 \times 10^{-27}$ cm^6/s.

The inset to Fig. 2 also emphasizes two complementary aspects of Feshbach resonances: When the field shifts a molecular state into resonance with the scattering continuum, an increase in radiative loss (lower curve) indicates the appearance of a molecular population, while a change in evaporation loss rate (upper curve) in the absence of the probe beam indicates a resonant alteration of the elastic collision cross section. The difference between these two effects is exemplified by the fact that the radiative resonance at 47.97 G occurs at a slightly lower field value than does the minimum in the evaporation loss at 48.02 G. While the radiative resonance occurs when the molecular state is tuned to the scattering continuum, the minimum in the elastic cross section occurs when the background scattering length is canceled by the resonant contribution from the bound state [9]. The finite width of the resonance causes these two points to be distinct.

The atomic (molecular) population $N(M)$ in thermal equilibrium for a purely elastic atom-molecule coupling can be calculated from the partition function $Z(N, M) = Z_n^N Z_m^M / N!M!$ subjected to the constraint $N + 2M = $ const, where Z_n (Z_m) is the partition function of a single atom (molecule). Maximizing $Z(N, M)$, we obtain $M/Z_m = N^2/Z_n^2$. In a box of volume V, the partition function is $Z = V/h^3 \lambda_{\mathrm{dB}}^3 e^{-E/k_B T}$, where E is the energy of the state, and $\lambda_{\mathrm{dB}} = h(2\pi M k_B T)^{-3/2}$ the thermal de Broglie wavelength of a particle of mass M at temperature T. The mean atomic density n and mean molecular density m are thus simply related by a Saha-Boltzmann–type equation [11] $m = n^2 \lambda_{\mathrm{dB}}^3 e^{-\Delta E/k_B T}$, where ΔE is the energy of the molecular state relative to the dissociation threshold. In a far-detuned optical trap, atoms and molecules experience a harmonic potential with a similar vibration frequency ω. Using the partition function for particles in a harmonic trap, $Z_m = Z_n = (1 - \hbar\omega/k_B T)^{-3}$, the relation between molecular and atomic density on the Feshbach resonance ($\Delta E = 0$) can be expressed in terms of the atomic peak phase space density $\phi = N/Z_n$ as $m = n\phi$. This simple result shows that in thermal equilibrium, and in the absence of any additional molecular loss processes, the ratio of molecular to atomic density is given by the phase space density of the atomic gas.

To model the dynamics of molecule formation, dissociation by collisions, and probe-beam-induced depletion in a thermal cloud, we adopt a rate equation approach that treats atoms and molecules separately [10,19]: Creation and depletion rates of molecules near a Feshbach resonance are modeled by αn^2 and $-\beta m$, respectively, where α and β characterize the coupling strength of the resonance. Given a leading order atom-molecule collision loss $-\gamma nm$, and radiative molecular loss induced by the probe beam $L/2 = \delta mI$, where γ and δ are rate constants, and I is the probe beam intensity, we obtain the following equations:

$$\dot{m} = \alpha n^2 - \beta m - \gamma nm - L/2, \tag{1}$$

$$L = 2\delta Im. \tag{2}$$

The stationary solutions for the molecular density m and the radiative loss rate constant of the atom gas L/n are then given by

$$m = \frac{\alpha n^2}{\beta + \gamma n + \delta I}, \tag{3}$$

$$\frac{L}{n} = \frac{2\delta \alpha In}{\beta + \gamma n + \delta I}. \tag{4}$$

To test this model, we measure the atom loss as a function of probe beam intensity I for different atomic densities on the 53.50 G resonance, shown in Fig. 3. Here the wavelength of the probe beam is 846.5 nm, and the mean atomic density is switched between $n_1 = 1.2 \times 10^{13}$ cm^{-3} and $n_2 = 0.6 \times 10^{13}$ cm^{-3} by changing the trap loading parameters. The phase space densities are $\phi_1 \approx 1/100$ and $\phi_2 \approx 1/200$, respectively. The radiative loss constant L/n is derived from the measured radiation exposure time t necessary to deplete 10% of the sample, namely, $L/n = 0.1/t$. The polarization purity of 95% in $|3, 3\rangle$ and the temperature of 5.5 μK do not vary systematically by more than 5% between the two settings. We
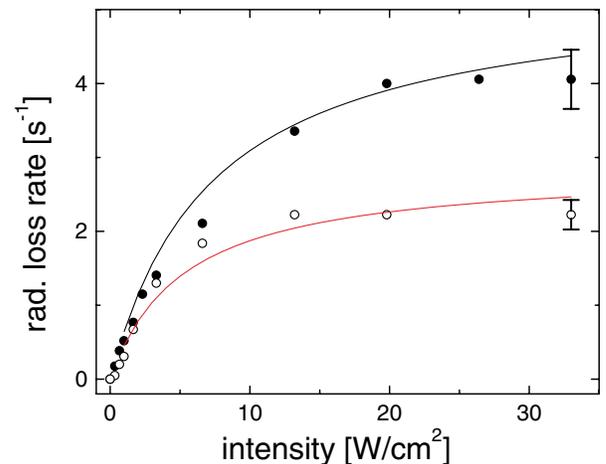

FIG. 3 (color online). Radiative loss rate L/n at the 53.50 G Feshbach resonance versus intensity for atomic densities of $n_1 = 1.2 \times 10^{13}$ cm^{-3} (solid circles) and $n_2 = 0.6 \times 10^{13}$ cm^{-3} at a temperature of 5.5 μK. We fit the two curves simultaneously with Eq. (4).

have verified that no observable loss is induced by field ramping, by the trapping laser or by the probe beam when the magnetic field is off resonant.

At low probe beam intensity $I < 5$ W/cm^2, the fractional loss rate is linear in I. This suggests that the molecular and atomic populations reach a dynamic equilibrium where the probe beam only weakly excites a small fraction of the molecules. At high probe beam intensity $I > 10$ W/cm^2, the fractional loss rate saturates to a constant value which is proportional to the sample density. This implies a quick depletion of molecular population by the probe beam, leading to a loss rate that is therefore equal to the formation rate of the molecules. As indicated by the solid lines in Fig. 3, Eq. (4) agrees well with the experimental results. A simultaneous fit to both sets of data yields $\alpha = 2.2(1) \times 10^{-13}$ cm^3 s^{-1}, the ratio of the rate constants $\delta/\gamma = 1.2(1) \times 10^{12}$ (W cm)$^{-1}$, and $\beta/\gamma < 1.5 \times 10^{12}$ cm^{-3}.

From the above measurements, we can deduce the sensitivity of our method, expressed in terms of a molecule formation cross section given by $\sigma_\alpha = \alpha/v$, where v is the mean relative velocity of the atoms at our temperature. From the data shown in Fig. 3, we obtain $\sigma_\alpha = 5 \times 10^{-14}$ cm^2. While this resonance induces a maximum loss rate of 4 s^{-1}, the loss rate of the weakest resonance at 7.8 G is approximately 0.04 s^{-1}, observed with a signal-to-noise ratio of 3. We then estimate that our minimum detectable molecule formation cross section is as small as $\sigma_\alpha/300 = 1.7 \times 10^{-16}$ cm^2.

Furthermore, we can estimate the stationary number of molecules in the gas from the measured rate constants. While the latter reveal only α and δ/γ, we can put an upper limit on the rate constant γ from the unitarity limit of the inelastic cross section: $\sigma_\gamma < \pi/k^2$. Here $\hbar^2 k^2/2\mu = \frac{3}{2} k_B T$ is the collision energy, $\mu = \frac{2}{3} M$ the reduced mass, and M the mass of the Cs atom. We then estimate $\gamma = \sigma_\gamma v' < (\pi/k^2)(\hbar k/\mu) = 4.1 \times 10^{-11}$ cm^3 s^{-1}. Given Eq. (3), α and $\beta/\gamma \lesssim 2 \times 10^{12}$ cm^{-3}, we obtain a ratio of molecular to atomic density $m/n > 0.005(1)$ at an atomic density of $n = 1.2 \times 10^{13}$ cm^{-3}, and low radiative excitation. Equivalently, more than $5(1) \times 10^5$ quasibound molecules coexist with 10^8 atoms in our trap at a molecular density m exceeding $6(1) \times 10^{10}$ cm^{-3}. Note that this fraction is below the ideal limit of $m = n\phi = 0.01n$.

In conclusion, we have used our radiative detection method to observe multiple Cs$_2$ molecular states with high sensitivity and resolution. Since no observable collision loss accompanies these resonances, as many as 5×10^5 quasibound molecules in a single rovibrational state accumulate and coexist dynamically with 10^8 atoms in our dipole trap. Although the molecular dimers we observe are only quasibound due to their coupling to the scattering continuum, it may be possible to convert them into longer-lived, bound molecules by dynamically tuning the molecular state below the scattering continuum on a time scale short compared to the inverse of the resonance width [10].

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