Observation of Feshbach-Like Resonances in Collisions between Ultracold Molecules

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(Received 10 November 2004; published 1 April 2005)

We observe magnetically tuned collision resonances for ultracold Cs_2 molecules stored in a CO_2-laser trap. By magnetically levitating the molecules against gravity, we precisely measure their magnetic moment. We find an avoided level crossing which allows us to transfer the molecules into another state. In the new state, two Feshbach-like collision resonances show up as strong inelastic loss features. We interpret these resonances as being induced by Cs_4 bound states near the molecular scattering continuum. The tunability of the interactions between molecules opens up novel applications such as controlled chemical reactions and synthesis of ultracold complex molecules.

DOI: 10.1103/PhysRevLett.94.123201 PACS numbers: 34.50.−s, 05.30.Jp, 32.80.Pj, 67.40.Hf

The synthesis of ultracold molecules from ultracold atoms has opened up new possibilities for studies on molecular matter waves [1–3], strongly interacting superfluids [4], high-precision molecular spectroscopy [5] and coherent molecular optics [6]. In all these experiments, control of the interatomic interaction by magnetic fields plays an essential role in the association process. When a two-atom bound state is magnetically tuned near the quantum state of two scattering atoms, coupling from the atomic to the molecular state can be resonantly enhanced. This is commonly referred to as a Feshbach resonance [7].

The success in controlling the interaction of ultracold atoms raises the question whether a similar level of control can be achieved for ultracold molecules. Resonant interactions between molecules may lead to synthesis of complex objects beyond atomic dimers. Furthermore, scattering processes for molecules involve many novel reactive channels in comparison to the atomic counterpart, e.g., collision induced dissociation, rearrangement, or displacement chemical reactions. Magnetic tunability of the molecular interactions, similar to that resulting from atomic Feshbach resonances, will lead to exciting perspectives for investigating these chemical processes in regimes where quantum statistics and quantum coherence play an important role.

In this Letter, we report the observation of magnetically tuned collision resonances in an ultracold gas of Cs_2 molecules. The ultracold dimers are created from an atomic Bose-Einstein condensate (BEC) by use of a Feshbach ramp [1] and are trapped in a CO_2-laser trap. We precisely measure the magnetic moment of the molecules and observe an avoided crossing [8] which allows us to transfer the molecules into another state. In the new state, we discover two narrow inelastic collision resonances. The resonance structure suggests that bound states of two cesium molecules, or equivalently Cs_4 states, induce the resonant scattering of molecules. These resonances, which we interpret as Feshbach resonances for ultracold molecules, may open the door to the synthesis of more complex molecules and to the control of their chemical reactions.

The relevant molecular energy structure shown in Fig. 1 is based on calculations done at NIST [9,10]. The dissociation threshold, providing the energy reference $E_b = 0$, is associated with two Cs atoms in the lowest ground state sublevel $|F = 3, m_F = 3\rangle$, where F and m_F are the quantum number of the atomic angular momentum and its projection, respectively. As a result of the strong indirect spin-spin interaction of Cs atoms [11], coupling to molecular states with large orbital angular momentum $l = 4$ [10,12] leads to the complexity of the energy structure shown in Fig. 1. This type of coupling is generally referred to as g-wave Feshbach coupling.

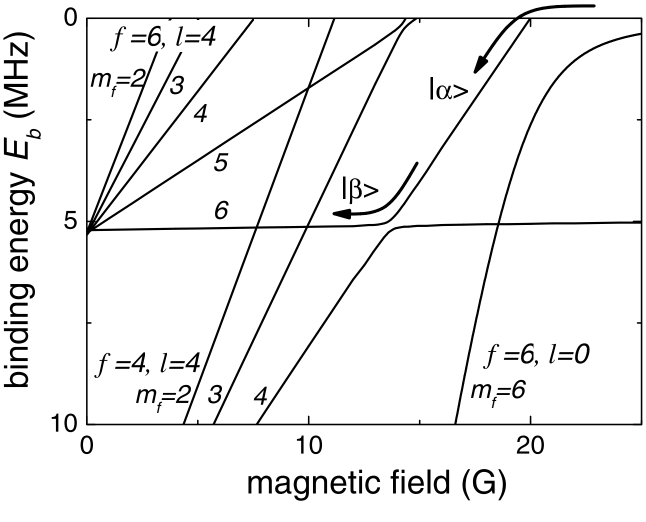

FIG. 1. Molecular energy structure below the scattering continuum of two cesium atoms in the $|F = 3, m_F = 3\rangle$ state. The energy of the dissociation threshold corresponds to $E_b = 0$. The arrows mark the paths to the molecular states we explore, which include the creation of the molecules in $|\alpha\rangle$ via the atomic Feshbach resonance at 19.84 G [1,19] and an avoided crossing to $|\beta\rangle$ at ~13.6 G. Included are only molecular states which can couple to the continuum via Feshbach couplings up to g-wave interaction ($l \leq 4$, $m_f + m_l = 6$ and $m_f \geq 2$).

We create the molecules in the bound state $|\alpha\rangle \equiv |f = 4, m_f = 4; l = 4, m_l = 2\rangle$ via g-wave Feshbach coupling at 19.84 G [1]; see Fig. 1. Here, f is the internal angular momentum of the molecule, and m_f and m_l are the projections of f and l, respectively. The molecular state $|\alpha\rangle$ is stable against spontaneous dissociation for magnetic fields below 19.84 G and acquires larger binding energies at lower magnetic fields. This is due to the small magnetic momentum of $\sim 0.95\mu_B$ of this state as compared to the atomic scattering continuum with $\sim 1.5\mu_B$. At about 14 G, an avoided crossing to another state $|\beta\rangle \equiv |f = 6, m_f = 6; l = 4, m_l = 0\rangle$ is induced by the indirect spin-spin coupling. In this work, we ramp the magnetic field adiabatically and explore the upper branch of the avoided crossing.

Our experiment starts with an essentially pure atomic BEC with up to 2.2×10^5 atoms in a crossed dipole trap formed by two CO_2 laser beams [13,14]. We apply a magnetic field of 20 G, slightly above the Feshbach resonance, and a magnetic field gradient of 31 G/cm to levitate the atoms [15]. The CO_2-laser trap is roughly spherically symmetric with a trapping frequency of $\omega \approx 2\pi \times 20$ Hz and a trap depth of 7 μK. The atomic density is 6×10^{13} cm^{-3} and the chemical potential is $k_B \times 20$ nK, where k_B is Boltzmann's constant.

To create the molecules, we first ramp the magnetic field from 20.0 to 19.5 G in 8 ms and then quickly change the field to 17 G to decouple the molecules from the atoms. Simultaneously, we ramp the magnetic field gradient from 31 up to 50 G/cm. The latter field gradient levitates the molecules [1] and removes all the atoms from the trap in 3 ms. As a consequence, we obtain a pure molecular sample in the CO_2-laser trap with typically 10^4 molecules. The magnetic field ramping process also leads to a small momentum kick on the molecules, which start oscillating in the trap. After $\sim$100 ms, the oscillations are damped out and the sample comes to a new equilibrium at a temperature of 250 nK with a peak density of 5×10^{10} cm^{-3} and a phase space density of 10^{-2} to 10^{-3}. To measure the molecule number, we dissociate the molecules into free atoms by reversely ramping the magnetic field back above the resonance to 21 G. We then image the resulting atoms [1].

A key parameter for a perfect levitation of the Cs_2 molecules is the precise value of their magnetic moment [1]. The levitation field is crucial because the gravitational force is much stronger than the trapping force of the CO_2 lasers. In contrast to ground state atoms with only slow-varying magnetic moment, the magnetic moment of the molecules can sensitively depend on the magnetic field as a result of the complex interactions between molecular states; see Fig. 1. Therefore, the prerequisite to perform Cs_2 molecule experiments at different magnetic fields is the knowledge of the molecular magnetic moment for an accurate setting of the levitation field.

We map out the magnetic moment of the molecules over the range of 11.5 to 19.8 G. This is realized by a two-step process: First, we slowly tune the magnetic field in 60 ms to a desired value and find a corresponding magnetic field gradient which can approximately keep the molecules near the center of the CO_2-laser trap. Second, after a hold time of 500 ms needed for the ensemble to come to an equilibrium, we measure the position of the cloud. The location of the molecular cloud provides a very sensitive probe to the residual imbalance of the magnetic force and gravity. Given a small vertical displacement of the molecules relative to the trap center δz for a local magnetic field B and a field gradient B', the magnetic moment is then $\mu(B) = (2m\omega^2\delta z + 2mg)/B'$. Here $2m$ is the molecular mass, and g is the gravitational acceleration. Independent measurements based on releasing the molecules into free space [1] confirm the accuracy of the above method to $0.01\mu_B$.

The measured magnetic moments of the molecules show the expected behavior in the range of 11.5 to 19.8 G; see Fig. 2. We find that the magnetic moment slowly decreases from $0.98\mu_B$ to $0.93\mu_B$ as the magnetic field is lowered from 19.8 G. For magnetic fields below $\sim$14 G, the magnetic moment quickly rises and levels off at $1.5\mu_B$. This behavior is readily explained by the avoided crossing at 13.6 G (Figs. 1 and 2), which transfers the molecules from state $|\alpha\rangle$ with $\mu \approx 0.9\mu_B$ to $|\beta\rangle$ with $\mu \approx 1.5\mu_B$. Below 11.5 G, a new avoided crossing to a very weakly coupled $l = 8$ molecular state occurs [16]. We observe fast loss of the molecules since our current apparatus cannot produce a sufficient levitation field to support the molecules against gravity in this new state.

Our measurement agrees excellently with the NIST calculation [9,10] within the 200 mG uncertainty from the multichannel calculation; see Fig. 2. We evaluate the

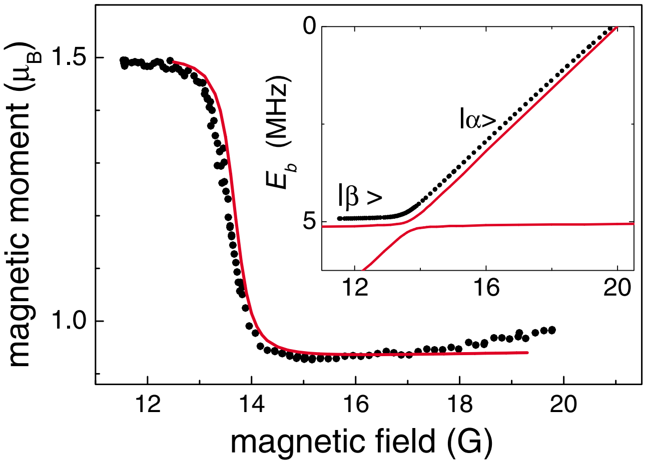

FIG. 2 (color online). Magnetic moment of the Cs_2 molecules. The measured magnetic moment (solid circles) is compared to the NIST calculation (solid line). The fast change at $\sim$13.6 G is associated with an avoided crossing. In the inset, we derive the molecular binding energy (solid circles) by integrating the measured magnetic moment. Binding energies from the NIST calculation (solid lines) for both branches of the avoided crossing between state $|\alpha\rangle$ and state $|\beta\rangle$ are shown; see also Fig. 1.

molecular binding energy based on integrating the measured magnetic moments. Here the integration constant is fixed by the fact that the molecular binding energy is zero at the atomic Feshbach resonance $B = 19.84$ G. The result shown in the inset of Fig. 2 gives very good agreement with the theoretical calculation within the energy uncertainty of 0.25 MHz [9]. By fitting our binding energies to a simple avoided crossing model, we determine the crossing to be $B_{\mathrm{cross}} = 13.55(4)$ G and the coupling strength, half the energy splitting between the two eigenstates at B_{cross}, to be $h \times 150(10)$ kHz. Here h is Planck's constant.

To investigate the interactions between molecules, we measure the inelastic collision loss after a trapping time of 300 ms (Fig. 3). For molecules in state $|\alpha\rangle$ ($14\ \mathrm{G} < B < 19.8\ \mathrm{G}$), the fractional loss is about ~40%. In this molecular state, we do not see any strong magnetic field dependence. When the magnetic field is tuned near the Feshbach resonance at 19.8 G, molecules dissociate into free atoms, which leave the trap.

In state $|\beta\rangle$ ($11.5\ \mathrm{G} < B < 13.6\ \mathrm{G}$), the behavior of the molecules is strikingly different. We observe a weaker background loss of ~20% and two pronounced resonances with a fractional loss of up to 60%. An expanded view in the inset of Fig. 3 shows that the "double peak" structure can be well fit by a sum of two Lorentzian profiles. From the fit, we determine the resonance positions to be 12.72(1) and 13.15(2) G with full widths of 0.25 and 0.24 G, respectively. Note that due to the levitation gradient field, the inhomogeneity across the molecular sample is as large as 0.15 G in state $|\beta\rangle$, which suggests that the intrinsic widths of these resonances are less than the observed values.

The observed resonances cannot be explained by single-molecule effects based on the Cs_2 energy structure, which is precisely known to very high partial waves [9,10]. Beyond single-molecule effects, the observed resonance structure strongly suggests that bound states of two Cs_2 molecules (Cs_4 tetramer states) are tuned in resonance with the scattering state of the molecules and induce Feshbach-like couplings to inelastic decay channels. Other possible scattering processes, e.g., direct coupling to a trimer and an atom or a dimer and two atoms, should result in a threshold behavior in the loss spectrum instead of the observed resonance structure. For Cs_2 molecules, the appearance of Cs_4 bound states near the scattering continuum is not surprising considering the complexity of interaction between Cs atoms and the additional rotational and vibrational degrees of freedom.

To confirm that the loss is indeed due to collisions between molecules, we observe the decay of the molecular population in the CO_2-laser trap. Starting with 11 000 molecules prepared at different magnetic fields, we record the molecule number after various wait times, as shown in Fig. 4. Three magnetic field values are chosen here: 15.4 G where the molecules are in state $|\alpha\rangle$, 12.1 G where the molecules are in state $|\beta\rangle$ and are away from the resonance, and 12.7 G where the molecules are on the strong molecular resonance; see Fig. 4. The number of trapped molecules shows a nonexponential decay, which provides a clear signature of the density-dependent processes.

To further investigate the underlying molecular collision processes, we model the loss based on a two-body or a three-body loss equation. Assuming a Gaussian distribution for the thermal ensemble in a harmonic trap with a constant temperature and that the collision loss rate is slow compared to the thermalization rate, we fit the measured molecule numbers to the two-body and three-body decay equation; see in Fig. 4. For 15.4 and 12.1 G, we find that the two-body equation provides excellent fits. The two-body

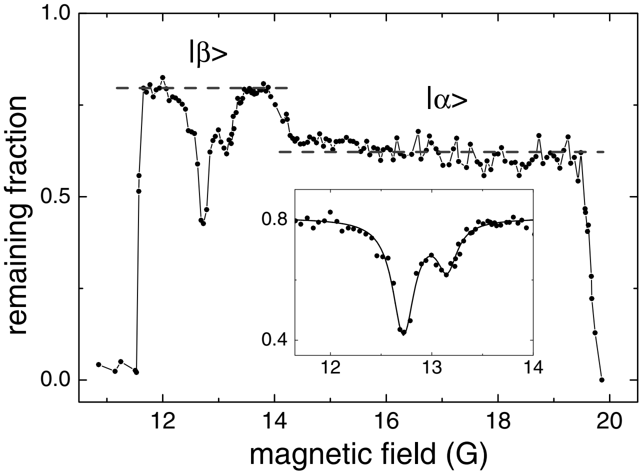

FIG. 3. Remaining fraction of optically trapped molecules after a storage time of 300 ms. Initially, there are 11 000 molecules at a peak density of 6×10^{10} cm^{-3} and a temperature of 250 nK. The dashed lines mark the background loss rates in state $|\alpha\rangle$ and in state $|\beta\rangle$. The two loss resonances for $|\beta\rangle$ are fit by a sum of two Lorentzian profiles (inset).

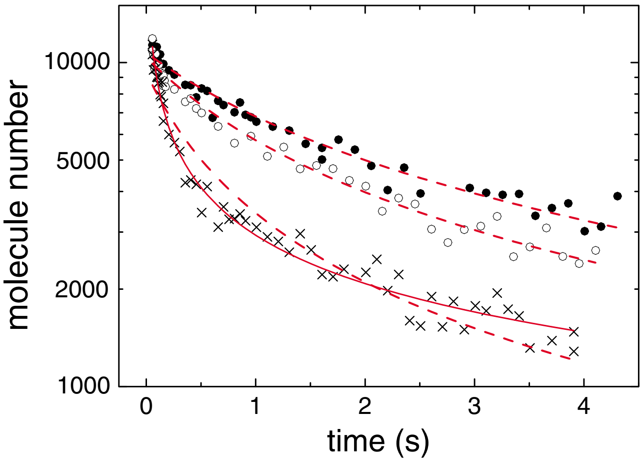

FIG. 4 (color online). Time evolution of the molecule number in the CO_2-laser trap for molecules in state $|\alpha\rangle$ at 15.4 G (open circles), in state $|\beta\rangle$ at 12.1 G (off resonance, solid circles) and at 12.7 G (on resonance, crosses). Fits based on two-body loss (dashed lines) work well for 15.4 and 12.1 G. A fit based on three-body loss (solid line) works better for 12.7 G.

coefficients are 5×10^{-11} cm^3/s at 15.4 G and 3×10^{-11} cm^3/s at 12.1 G. We cannot, however, rule out the possibility that three-body processes also play a role. The measured collision rate coefficients are similar to the measurements from the MIT group on Na_2 [17], and are an order of magnitude below the unitarity limit of $2h/mk = 4 \times 10^{-10}$ cm^3/s, where k is the characteristic collision wave number associated with the temperature of the sample.

At 12.7 G, where the molecules are on the strong resonance, we find that the three-body equation actually provides a better fit than the two-body fit with a three-body loss coefficient at 6×10^{-20} cm^6/s; see Fig. 4. This value, however, is much too high compared to the three-body unitarity limit of $96\pi h/mk^4 = 2 \times 10^{-23}$ cm^6/s [18]. One alternative explanation is that on resonance, the fast collision loss rate might leave the molecules insufficient time to reach thermal equilibrium. By fitting the resonance data in the first 200 ms with the two-body loss model, we determine the two-body loss coefficient to be 2×10^{-10} cm^3/s, which indeed approaches the unitarity limit of 4×10^{-10} cm^3/s.

In conclusion, we have observed magnetically tuned collision resonances in a trapped ultracold sample of Cs_2 dimers. The density-dependent inelastic decay and the resonance structure strongly suggest a resonant coupling to Cs_4 tetramer states. Our observations are reminiscent of Feshbach resonances in atom-atom scattering. The controlled use of such resonances for interaction tuning and molecule formation in atomic ensembles has opened up new avenues in research on ultracold quantum gases. Our observation of magnetically tuned Feshbach-like resonances in molecule-molecule scattering brings in fascinating prospects for a controlled synthesis of ultracold tetramers in a single four-body quantum state in analogy to the formation of ultracold dimers near atomic Feshbach resonances. The tunability of the interactions in molecular quantum gases can potentially open up the door to few-body physics beyond simple atoms and diatomic molecules and to a new ultracold chemistry.

We greatly thank E. Tiesinga and P. S. Julienne for stimulating discussions, and, in particular, for providing us with the theoretical calculation on the Cs_2 energy structure. We acknowledge support by the Austrian Science Fund (FWF) within SFB 15 and the Lise Meitner program, and by the European Union in the frame of the Cold Molecules TMR Network under Contract No. HPRN-CT-2002-00290. M. M. is supported by DOC [Doktorandenprogramm der Österreichischen Akademie der Wissenschaften].

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