Observation of Feshbach-like resonances in collisions between ultracold molecules

C. Chin,1 T. Kraemer,1 M. Mark,1 J. Herbig,1 P. Waldburger,1 H.-C. Nägerl,1 R. Grimm1,2
1Institut für Experimentalphysik, Universität Innsbruck, Technikerstraße 25, 6020 Innsbruck, Austria
2Institut für Quantenoptik und Quanteninformation, Österreichische Akademie der Wissenschaften, 6020 Innsbruck, Austria
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We observe magnetically tuned collision resonances for ultracold Cs2 molecules stored in a CO2-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 Cs4 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.

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The synthesis of ultracold molecules from ultracold atoms has opened up new possibilities for studies on molecular matter-waves, strongly interacting superfluids, high-precision molecular spectroscopy and coherent molecular optics. 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.

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 to investigate 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 Cs2 molecules. The ultracold dimers are created from an atomic Bose-Einstein condensate (BEC) by use of a Feshbach ramp and trapped in a CO2-laser trap. We precisely measure the magnetic moment of the molecules and observe an avoided crossing 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 Cs4 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. is based on calculations done at NIST. The dissociation threshold, providing the energy reference $E_0 = 0$, 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, coupling to molecular states with large orbital angular momentum $l = 4$ leads to the complexity of the energy structure shown in Fig. 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 $E_0 = 0$. 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 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 $2.2 \times 10^5$ atoms in a crossed dipole trap formed by two CO$_2$ laser beams [14,15]. We apply a magnetic field of 20 G, slightly above the Feshbach resonance, and a magnetic field gradient of 31.2 G/cm to levitate the atoms [16]. 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 $\mu$K. The atomic density is $6 \times 10^{13}$ 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 G 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 31.2 up to 50.2 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 $5 \times 10^{10}$ 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 atom number [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 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 G 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 $\delta z$ 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 G 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 (Fig. 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 [17]. 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 [10,11] within the 200 mG uncertainty from the multi-channel calculation, see Fig. 2. We evaluate the
states at strength, half the energy splitting between the two eigen-
energies to a simple avoided crossing model, we determine the uncer-
tainty of 0.25 MHz [10]. By fitting our binding en-
result shown in the inset of Fig. 2 gives very good agree-
Because of the gradients, the inhomogeneity across the molecular}
structure can be well fit by a sum of two Lorentzian pro-
ances with a fractional loss of up to 60%. An expanded view in the inset of Fig. 3 shows that the “double peak”
are fit by a sum of two Lorentzian profiles (inset).

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 G < $B < 19.8$ 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 G < $B < 13.6$ 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) G and 13.15(2) G with full widths of 0.25 G 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 struc-
ture, which is precisely known to very high partial waves [10, 11]. Beyond single-molecule effects, the observed resonance structure strongly suggests that bound states of Cs$_2$ molecules (Cs$_4$ states) are tuned in resonance with the scattering state of the molecules and induce Feshbach-like couplings to inelastic decay channels. 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 verify that the loss is indeed due to collisions between molecules, we measure the time evolution of the molecular population in the CO$_2$-laser trap. Starting with 11000 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 non-
exponential decay, which provides a clear signature of density-dependent collision processes. To determine the underlying 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- and three-body decay equation, see in Fig. 4.

For 15.4 G and 12.1 G, we find that the two-body equation provides excellent fits. The two-body coefficients are $5 \times 10^{-11}$ cm$^3$/s at 15.4 G and $3 \times 10^{-11}$ cm$^3$/s at 12.1 G. We, however, cannot 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$_2$ [12], and are an order of magnitude below the unitarity limit of $2 \hbar/mk = 4 \times 10^{-10}$ 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 $6 \times 10^{-20}$ cm$^3$/s, see Fig. 4. This value, however, is much too high compared to the three-body unitarity limit of $96\pi \hbar/mk^3 = 2 \times 10^{-13}$ cm$^3$/s [13]. 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 $2 \times 10^{-10}$ cm$^3$/s, which indeed approaches the unitarity limit of $4 \times 10^{-10}$ cm$^3$/s.

The roles of two-body and three-body processes near a collision resonance have been extensively studied for ultracold atoms. For two-body losses, colliding atoms can only decay into two free atoms with lower internal energies. For three-body losses, the outgoing channels,
in general, are either three free atoms or one atom and one dimer. For molecules, however, many more outgoing channels are possible. For instance, two resonantly interacting Cs₂ molecules can decay into four Cs atoms, two Cs atoms and one Cs₂ dimer, or one Cs atom and one Cs₃ trimer. In the case of three-molecule collisions, there exist, in principle, ten different types of decay processes.

More intriguingly, the observed magnetically tuned Feshbach-like resonances bring in fascinating prospects for a controlled synthesis of Cs₄ tetramers in a single four-body bound state. This is analogous to the formation of the Cs₂ dimers near the atomic Feshbach resonances. The tunability of the interactions in molecular quantum gases can potentially open up the door to ultracold chemistry and to few-body physics beyond simple atoms and diatomic molecules.

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[1] J. Herbig, T. Kraemer, M. Mark, T. Weber, C. Chin, H.-C. Nägerl, and R. Grimm, Science 301, 1510 (2003).
[2] K. Xu, T. Mukaiyama, J. R. Abo-Shaeer, J. K. Chin, D. E. Miller, and W. Ketterle, Phys. Rev. Lett. 91, 210402 (2003).
[3] S. Jochim, M. Bartenstein, A. Altmeyer, G. Hendl, S. Riedl, C. Chin, J. Hecker Denschlag and R. Grimm, Science 302, 2101 (2003); M. Greiner, C.A. Regal, D.S. Jin, Nature 426, 537 (2003); Zwerlein, C.A. Stan, C.H. Schunck, S.M.F. Raupach, S. Gupta, Z. Hadzibabic, and W. Ketterle, Phys. Rev. Lett. 91, 250401 (2003).
[4] M. Bartenstein, A. Altmeyer, S. Riedl, S. Jochim, C. Chin, J. Hecker Denschlag and R. Grimm, Phys. Rev. Lett. 92, 120401 (2004); C.A. Regal, M. Greiner, D.S. Jin, Phys. Rev. Lett. 92, 040403 (2004); M.W. Zwerlein, C.A. Stan, C.H. Schunck, S.M. F. Raupach, A.J. Kerman, and W. Ketterle, Phys. Rev. Lett. 92, 120403 (2004); T. Bourdel, L. Khaykovich, J. Cubizolles, J. Zhang, F. Chevy, M. Teichmann, L. Tarruell, S.J.J.M.F. Kokkelmans, and C. Salomon Phys. Rev. Lett. 93, 050401 (2004).
[5] M. Bartenstein, A. Altmeyer, S. Riedl, R. Geursen, S. Jochim, C. Chin, J. Hecker Denschlag, R. Grimm, A. Simoni, E. Tiesinga, C.J. Williams, P.S. Julienne, cond-mat/0408673
[6] J.R. Abo-Shaeer, D.E. Miller, J.K. Chin, K. Xu, T. Mukaiyama, W. Ketterle, cond-mat/0409327
[7] E. Tiesinga, B.J. Verhaar, and H.T.C. Stoof, Phys. Rev. A 47, 4114 (1993); S. Inouye, M. Andrews, J. Stenger, H.-J. Miesner, S. Stamper-Kurn, and W. Ketterle, Nature 392, 151 (1998).
[8] S. Dürr, T. Volz, A. Marte, and G. Rempe, Phys. Rev. Lett. 92, 020406 (2004).
[9] M. Mark, T. Kraemer, J. Herbig, C. Chin, H.-C. Nägerl, R. Grimm, cond-mat/0409737
[10] E. Tiesinga and P. S. Julienne, private communication.
[11] C. Chin, V. Vuletić, A.J. Kerman, S. Chu, E. Tiesinga, P. Leo, and C.J. Williams, Phys. Rev. A 70, 032701 (2004).
[12] F.H. Mies, C.J. Williams, P.S. Julienne, and M. Krauss, J. Res. Natl. Inst. Stand. Technol. 101, 521 (1996).
[13] C. Chin, V. Vuletić, A.J. Kerman, and S. Chu, Phys. Rev. Lett. 90, 033201 (2003).
[14] T. Kraemer, J. Herbig, M. Mark, T. Weber, C. Chin, H.-C. Nägerl, R. Grimm, Appl. Phys. B (in press), cond-mat/0408268
[15] We adiabatically transfer the atomic BEC from a near-infrared laser trap, where the atomic BEC is formed in the CO₂-laser trap by simultaneously reducing the near-infrared laser power to zero and ramping the CO₂ lasers to full power in 1 s.
[16] T. Weber, J. Herbig, M. Mark, H.-C. Nägerl, and R. Grimm, Science 299, 232 (2003).
[17] This I = 8 state, not shown in Fig. 1, is very weakly coupled to the state |β⟩. Investigations on this state are currently in progress.
[18] T. Mukaiyama, J.R. Abo-Shaeer, K. Xu, J.K. Chin, and W. Ketterle, Phys. Rev. Lett. 92, 180402 (2004).
[19] H. Suno, B.D. Esry, and C.H. Greene, Phys. Rev. Lett. 90, 053202 (2003).