CHEMICAL PHYSICS

Observation of magnetically tunable Feshbach resonances in ultracold ^{23}Na^{40}K + ^{40}K collisions

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Resonances in ultracold collisions involving heavy molecules are difficult to simulate theoretically and have proven challenging to detect. Here we report the observation of magnetically tunable Feshbach resonances in ultracold collisions between potassium-40 (^{40}K) atoms and sodium-23–potassium-40 (^{23}Na^{40}K) molecules in the rovibrational ground state. We prepare the atoms and molecules in various hyperfine levels of their ground states and observe the loss of molecules as a function of the magnetic field. The atom-molecule Feshbach resonances are identified by observing an enhancement of the loss. We have observed 11 resonances in the magnetic field range of 43 to 120 gauss. The observed atom-molecule Feshbach resonances at ultralow temperatures probe the three-body potential energy surface with exceptional resolution and will help to improve understanding of ultracold collisions.

Understanding collisions involving molecules at the quantum level has been a long-standing goal in chemical physics (*1*). Scattering resonances are among the most remarkable quantum phenomena and play a critically important role in the study of collisions. They are sensitive to both the long-range and short-range portions of the molecule interaction potential and thus offer an ideal probe of the potential energy surface (PES) governing the collision dynamics. In theory, describing the PES requires solving the Schrödinger equation involving many electrons and nuclei, which is notoriously difficult owing to the electron correlations. Therefore, measurement of scattering resonances not only provides a global and accurate probe of the PES but also helps provide understanding of the complicated quantum many-body problem.

Although scattering resonances are well known and have been the main features studied in ultracold atomic gases and nuclear collisions (*2*), they have proven challenging to observe in molecular systems. Recently, major progress has been achieved in the experimental study of resonances in cold molecular collisions involving light particles—for example, H_2, HD molecules, or He atoms—by means of molecular beam techniques. In crossed-beam or merged-beam experiments, shape resonances or Feshbach resonances have been observed in atom-molecule chemical reactions (*3*–*8*), atom-molecule inelastic collisions (*9*–*11*), and molecule-molecule inelastic collisions (*12*, *13*). However, in these experiments, the collision energies are still high (at kelvin or subkelvin), and thus a few partial waves contribute to the scattering cross sections.

Ultracold molecules offer great opportunities to study molecular collisions in the quantum regime. At ultralow temperatures, the de Broglie wavelength of the collision partners is much larger than the range of molecular interaction potential, and only the lowest possible partial wave of relative orbital angular momentum dominates the collision process (*14*, *15*). Consequently, the collisions at ultracold temperatures are highly quantum mechanical. Owing to the anisotropy of the PES, the collisions involving ultracold molecules may support many resonances that are contributed by the rotational and vibrational excited states (*16*, *17*). Therefore, it is expected that scattering resonances should be routinely observed in ultracold molecular systems. For ultracold collisions involving light molecules, the low density of resonant states allows calculations of the scattering resonances, and many Feshbach resonances in atom-molecule collisions (*18*–*20*) and molecule-molecule collisions (*16*, *21*) have been predicted. However, owing to the experimental difficulties of preparing the ultracold colliding particles, these predictions have not been tested.

The situation is much more complicated for ultracold collisions involving heavy molecules, such as the alkali-metal-diatomic molecules in the rovibrational ground state created from ultracold atomic gases (*22*–*28*). The scattering resonances involving these heavy molecules are difficult to calculate and are highly challenging to observe. For reactive collisions, the reactions are universal, and the short-range losses with a near-unity probability suppress any possible resonances (*15*, *29*). For nonreactive atom-molecule collisions, the PES is so deep that thousands of rovibrational states may contribute to the resonances. As a consequence, the density of resonant states near the threshold of the collision channel is quite high, and it is not clear whether the individual resonances are resolvable (*17*). In this case, the theoretical calculation of the Feshbach resonances is extremely difficult, especially when nuclear spins and external fields are considered (*30*). Instead, a statistical model has been adopted to explore such highly resonant scattering (*17*), which predicts that at a temperature below 1 μK, for atom-diatomic-molecule collisions, many s-wave Feshbach resonances with an average spacing of less than 1 gauss should be observable. However, the experimental observation of these resonances remains elusive.

Here we report the observation of magnetic Feshbach resonances between ultracold ^{23}Na^{40}K ground-state molecules and ^{40}K atoms. The binding energies of ^{23}Na^{40}K [vibrational quantum number (v) = 0] and $^{40}K_2$ ($v = 0$) are about 5212 cm^{-1} (*25*) and 4405 cm^{-1} (*29*), respectively. Therefore, ^{23}Na^{40}K (v = 0, N = 0) + ^{40}K collisions with N the rotational quantum number are nonreactive: The reaction $^{23}\mathrm{Na}^{40}\mathrm{K}\,(v=0) + {}^{40}\mathrm{K} \rightarrow {}^{40}\mathrm{K}_2\,(v=0) + {}^{23}\mathrm{Na}$ is highly endothermic and is forbidden at ultracold temperatures. The atomization energy of NaK_2 is estimated to be 7125 cm^{-1} (*31*), which gives rise to a deep PES. As illustrated in Fig. 1, the channels that are asymptotically closed support many triatomic bound states, which may lead to a high density of resonant states near the threshold. We prepared ^{23}Na^{40}K molecules and ^{40}K atoms in various hyperfine levels of their ground states and searched for the resonances by measuring the loss rate of the molecules due to atom-molecule inelastic collisions as a function of the magnetic field. The appearance of a Feshbach resonance is identified by observing a resonantly enhanced loss. We have observed 11 resonances in the magnetic field range $43 < B < 120$ G, where B is the magnetic field.

We first searched for the atom-molecule Feshbach resonances in the magnetic field range $99.3 < B < 103.8$ G, which is close to a broad atomic Feshbach resonance at 110 G. We created weakly bound Feshbach molecules in an ultracold ^{23}Na and ^{40}K atomic mixture at a temperature of about 500 nK by Raman photoassociation. The remaining ^{23}Na atoms were removed immediately after the Feshbach molecules were formed. We then transferred the molecules from the Feshbach state to the rovibrational ground state by means of stimulated Raman adiabatic passage (STIRAP). The details of the association and the STIRAP are given in the supplementary materials (*32*). The hyperfine levels of the ground states of the ^{23}Na^{40}K molecule are labeled by $|v, N, m_{I_{\mathrm{Na}}}, m_{I_{\mathrm{K}}}\rangle$, where the vibrational and rotational quantum numbers are $v = N = 0$ and $m_{I_{\mathrm{Na}}}$

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and m_{I_K} are the nuclear spin projections of ^{23}Na and ^{40}K, respectively. In our experiment, the hyperfine states $|0,0,-3/2,-2\rangle$, $|0,0,-3/2,-1\rangle$, $|0,0,-1/2,-3\rangle$, and $|0,0,-1/2,-2\rangle$ could be populated by choosing proper intermediate states and laser polarizations. The hyperfine structure of the ground-state molecule is shown in Fig. 1. After the ground-state molecules were prepared, the ^{40}K atoms were transferred to different hyperfine states $|f,m_f\rangle_K$ by radio frequency pulses, with the atomic angular momentum $f = 9/2$ and the projection quantum number $m_f = -9/2, ..., -1/2$. In this way, 20 different combinations of the atom and molecule hyperfine states could be prepared.

The ^{23}Na^{40}K molecules decay owing to two-body hyperfine-changing inelastic collisions with the ^{40}K atoms because the atoms and molecules are in excited hyperfine states. The hyperfine change may be caused by the interaction between the nuclear spins of the molecules and the unpaired electron spin of the atoms during the collision process (*17*). We recorded the time evolution of the number of the molecules, as shown in Fig. 2. After a certain hold time, the number of the remaining ground-state molecules was measured by transferring the molecules back to the Feshbach states, which were detected by absorption imaging. The typical lifetime of the molecules in the atom-molecule mixture is on the order of 10 ms. This is much shorter than the lifetime of the pure molecule gas, which is longer than 100 ms in the whole magnetic field window. Therefore, the decay of the molecule in the mixture is dominantly caused by atom-molecule inelastic collisions.

The decay of the molecules may be described by $dN_m/dt = -\gamma N_m$, where N_m is the number of molecules, t is time, and $\gamma = \beta\bar{n}_a$ is the decay rate, with β and $\bar{n}_a$ being the loss rate coefficient and the mean density of the ^{40}K atoms, respectively. The mean atomic density may be calculated by $\bar{n}_a = [(m_K\bar{\omega}^2)/(4\pi k_B T_K)]^{3/2} N_a$, where $\bar{\omega}$ is the geometric mean trapping frequencies of the ^{40}K atoms, k_B is the Boltzmann constant, T_K is the temperature, and N_a is the number of ^{40}K atoms. In our experiment, the number of ^{40}K atoms is about one order of magnitude larger than that of the molecules, and thus the mean density $\bar{n}_a$ is approximately a constant. In this case, the loss rate coefficient β may be extracted from the measured decay rate γ and the atomic mean density $\bar{n}_a$.

We searched for the atom-molecule Feshbach resonances in 20 different incoming collision channels. For each channel, we measured the loss rate coefficient as a function of the magnetic field. By varying the magnetic field, we expected to change the energy differences between the triatomic bound states and the threshold of the incoming scattering channel. If a triatomic bound state intersects the threshold of the scattering channel and the coupling between the bound state and the scattering state is strong, a Feshbach resonance may occur. The Feshbach resonances are identified through the strongly enhanced loss rate coefficients (*2*, *17*).

In the experiment, we found that the loss rate coefficients were different for various collision channels. For each channel, in most cases, the loss rate coefficients did not change considerably in the magnetic field range (*32*). However, in the $|0,0,-3/2,-2\rangle + |9/2,-3/2\rangle$, $|0,0,-3/2,-2\rangle + |9/2,-7/2\rangle$, and $|0,0,-1/2,-3\rangle + |9/2,-7/2\rangle$ collision channels, the loss rate coefficients show prominent features at about 101 G (Fig. 3). We attribute these loss features to the resonant enhancement of the inelastic collisions due to the s-wave atom-molecule Feshbach resonance. The resonance positions and widths obtained by the Gaussian fits are listed in Table 1.

It is valuable to compare the measured loss rate coefficients with the universal rate coefficient (*15*, *29*), which assumes short-range loss with unity probability. Using the parameters in (*17*), the s-wave universal rate coefficient is estimated to be about 1.3 × 10^{-10} cm^3/s. The background loss rate coefficients are usually smaller than the universal rate coefficient, except in the $|0,0,-1/2,-3\rangle + |9/2,-5/2\rangle$ channel. The resonantly enhanced loss rate coefficients in the three collision channels are larger than the universal rate coefficient by a factor of about 2 to 3.

We used a similar method to measure the loss rate coefficients in the magnetic field range of 89.4 to 89.9 G, close to an atomic Feshbach resonance at about 90.3 G (*32*). In this magnetic field window, we found that for the $|0,0,-3/2,-2\rangle + |9/2,-9/2\rangle$ channel, the loss rate coefficients in the range of 89.4 to 89.9 G are notably larger than the coefficients in the range of 99.3 to 103.8 G, which indicates that a loss feature may exist near 90 G. We also performed similar measurements in the magnetic field range of 84.4 to 85.6 G, where the atom and molecule can both be prepared in the lowest hyperfine states. In this

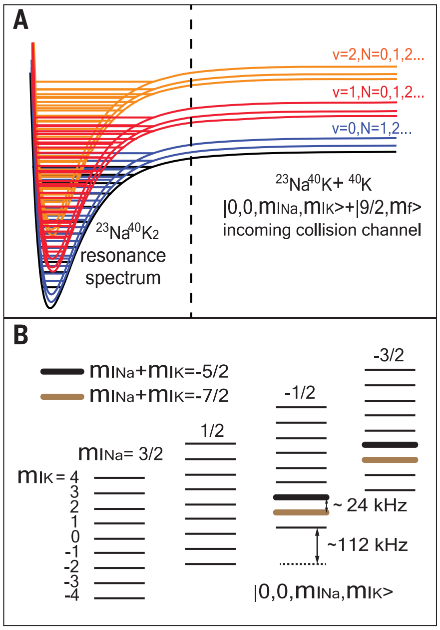

Fig. 1. Illustration of the atom-molecule Feshbach resonances between the ground-state ^{23}Na^{40}K molecule and ^{40}K atom. (**A**) The PES is very deep, and thus a large number of channels that are asymptotically closed can support the triatomic bound states, which give rise to a high density of resonant states near the threshold. The incoming channel is ^{23}Na^{40}K (v = 0, N = 0) + ^{40}K in a specific combination of hyperfine states. The atom-molecule Feshbach resonances probe the short-range resonance spectrum. The energy of the collision channels can be magnetically tuned. A Feshbach resonance occurs once the energy of the incoming channel coincides with the energy of a bound state. (**B**) Hyperfine structure of the ^{23}Na^{40}K ground-state molecule at a magnetic field of 100 G. The hyperfine levels of the ^{23}Na^{40}K molecule in the rovibrational ground state of the $^1\Sigma$ singlet potential are split owing to the nuclear Zeeman effects. The nuclear spin projections $m_{I_{Na}}$ and m_{I_K} are approximately good quantum numbers. The hyperfine levels that are used in the experiment are marked by thick black and brown lines.

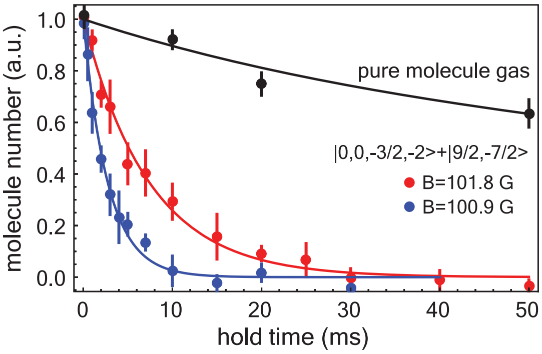

Fig. 2. The decay of the ^{23}Na^{40}K molecule in the atom-molecule mixture. The time evolutions of the number of the molecules are recorded. The solid curves are exponential fits with reduced chi-square values of 0.86 (red line) and 2.8 (blue line). The fits are not weighted to error bars. The loss rate coefficients are extracted from the measured decay rate. As a reference, the decay of the pure molecule gas in the $|0,0,-3/2,-2\rangle$ state at a magnetic field of 100.9 G is also shown. For the $|0,0,-3/2,-2\rangle + |9/2,-7/2\rangle$ collision, it can be clearly seen that the loss rates are dependent on the magnetic field. Each data point represents the average of three to five measurements, and the error bars represent 1 SD of the molecule number. a.u., arbitrary units.

magnetic field range, we did not observe an enhanced loss feature. We might expect that preparing both the molecule and atom in the lowest hyperfine states would largely suppress the loss rate. However, compared with the case in which the atom is prepared in the $|9/2, -7/2\rangle$ state, we do not observe a notable suppression of the loss rate. More theoretical and experimental studies are needed to understand these loss rate coefficients.

The observation of the three resonant loss features indicates that the resonance is resolvable. However, the magnetic field range that can be studied is limited close to the atomic Feshbach resonance, because the experiments are performed at fixed magnetic fields. This has hindered us from locating the possible resonance between $|0, 0, -3/2, -2\rangle$ and $|9/2, -9/2\rangle$ at about 90 G. To observe more resonances, the magnetic field range was expanded to $43 < B < 120$ G as follows. We first prepared the atom-molecule mixture at 102.3 G. After that, we swept the magnetic field to a desired strength in 2.5 to 4 ms. We have used the pre-emphasis method (*32*–*34*) to compensate for the magnetic fields created by the eddy currents induced by the stainless chamber or large coils. The atom-molecule mixture was held at the desired magnetic field for about 7 ms. During the hold time, the realistic magnetic field was within 100 to 400 mG of the desired magnetic field. The hold time was chosen in such a way that the resonantly enhanced loss could be clearly distinguished from the background loss. The ^{40}K atoms were then removed and the magnetic field swept back to 102.3 G in 3 ms, where the remaining molecules were transferred back to Feshbach state for detection. The Feshbach resonances manifest as the loss features of the remaining molecule number versus the magnetic field. Using this method, we have studied the collisions between the molecule state $|0, 0, -3/2, -2\rangle$ and the atom states $|9/2, -9/2\rangle$, $|9/2, -7/2\rangle$, and $|9/2, -5/2\rangle$, with a step of about 0.5 G. As shown in Fig. 4, we have observed eight new resonantly enhanced loss features. The resonance positions and widths obtained by the Gaussian fits are listed in Table 1. The resonance between $|0, 0, -3/2, -2\rangle$ and $|9/2, -9/2\rangle$ at about 90 G is clearly localized. The resonance between $|0, 0, -3/2, -2\rangle$ and $|9/2, -7/2\rangle$ at 101 G is also observed with this method.

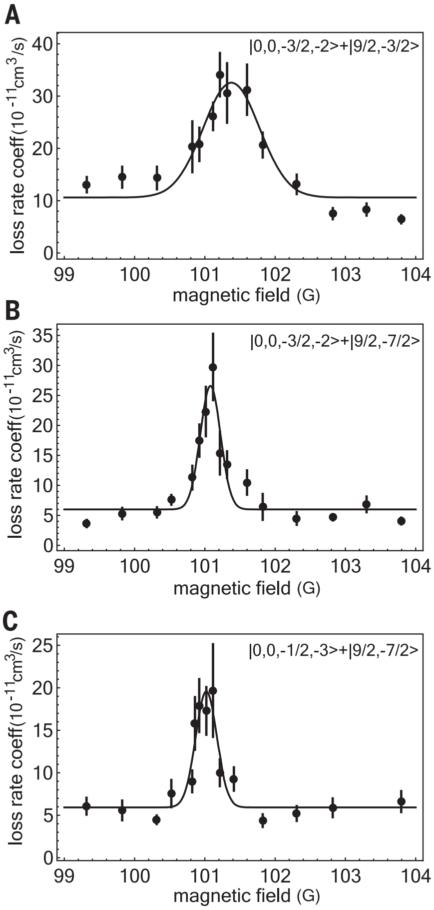

Fig. 3. Observations of the atom-molecule Feshbach resonances in the loss rate coefficients. (**A** to **C**) The loss rate coefficients are plotted as a function of the magnetic field. The collision channels are $|0, 0, -3/2, -2\rangle + |9/2, -3/2\rangle$ (A), $|0, 0, -3/2, -2\rangle + |9/2, -7/2\rangle$ (B), and $|0, 0, -1/2, -3\rangle + |9/2, -7/2\rangle$ (C). For these three channels, the resonantly enhanced loss rate coefficients at about 101 G provide clear evidence of the atom-molecule Feshbach resonances. The solid lines are phenomenological Gaussian fits with reduced chi-square values of 5.4 (A), 4.2 (B), and 2.4 (C). The fits are not weighted to error bars. The error bars represent 1 SD of the loss rate coefficients arising from the fitting uncertainty of decay rates and uncertainty of atomic densities.

The observation of the Feshbach resonances allows us to compare these values with the density of resonant states estimated from the statistical model. For the ^{23}Na^{40}K + ^{40}K collision studied in our experiment, neglecting the nuclear spins, the density of resonant states is estimated to be about 1.22 per mK (*17*). If the nuclear spins are considered, the density of resonant states is multiplied by the number of spin states that conserve the total magnetic quantum number. Assuming that the short-range physics does not change with the magnetic field, the resonance spectrum is probed with a rate of the Zeeman shift of the scattering channel (*17*). These arguments predict many s-wave resonances with an average spacing of about 1 G. However, in the approximate 70-G-wide magnetic field range, we observe only 11 resonantly enhanced loss features. This indicates that the density of resonant states may be not as large as the statistical model predicts. We cannot exclude the possibility that there are some narrow resonances that are not observed in our experiment.

In conclusion, we have observed magnetically tunable Feshbach resonances in ultracold collisions between ^{23}Na^{40}K ground-state molecules and ^{40}K atoms. In such a heavy and ultracold system, there may be many resonances in a magnetic field range of a few hundred gauss. The observation of more resonances may enable the study of the quantum chaos in ultracold molecular collisions (*17*).

The observed ultracold atom-molecule scattering resonances probe the short-range resonance spectrum with exceptional resolution and provide valuable information about the PES. So far, the accuracy of the PES calculated by solving the electronic Schrödinger equation is on the order of cm^{-1}, which is too low to be used to quantitatively understand these resonances. Therefore, the experimental observation of ultracold atom-molecule resonances challenges the accuracy of quantum chemistry simulations. In this sense, the observation of ultracold resonances provides a well-controlled and powerful tool to accurately simulate the quantum many-body problem in quantum chemistry. The observation of Feshbach resonances also opens up

Table 1. The Feshbach resonance position B_0 and width ΔB obtained by the Gaussian fits. The first three resonances are observed by measuring the loss rates at fixed magnetic fields. The other resonances are observed by sweeping the magnetic field. The resonance between $|0, 0, -3/2, -2\rangle$ and $|9/2, -7/2\rangle$ at 101 G is observed by both methods, and the second method gives a larger width.

Method	Collision channel	B_0 (G)	ΔB (G)
I	$\|0, 0, -3/2, -2\rangle + \|9/2, -3/2\rangle$	101.4	0.6
	$\|0, 0, -3/2, -2\rangle + \|9/2, -7/2\rangle$	101.1	0.2
	$\|0, 0, -1/2, -3\rangle + \|9/2, -7/2\rangle$	101.0	0.2
II	$\|0, 0, -3/2, -2\rangle + \|9/2, -5/2\rangle$	68.0	1.6
		74.2	3.5
		83.2	2.0
	$\|0, 0, -3/2, -2\rangle + \|9/2, -7/2\rangle$	54.5	0.6
		59.1	3.7
		101.0	0.6
		106.7	1.7
	$\|0, 0, -3/2, -2\rangle + \|9/2, -9/2\rangle$	48.1	2.6
		89.8	2.9

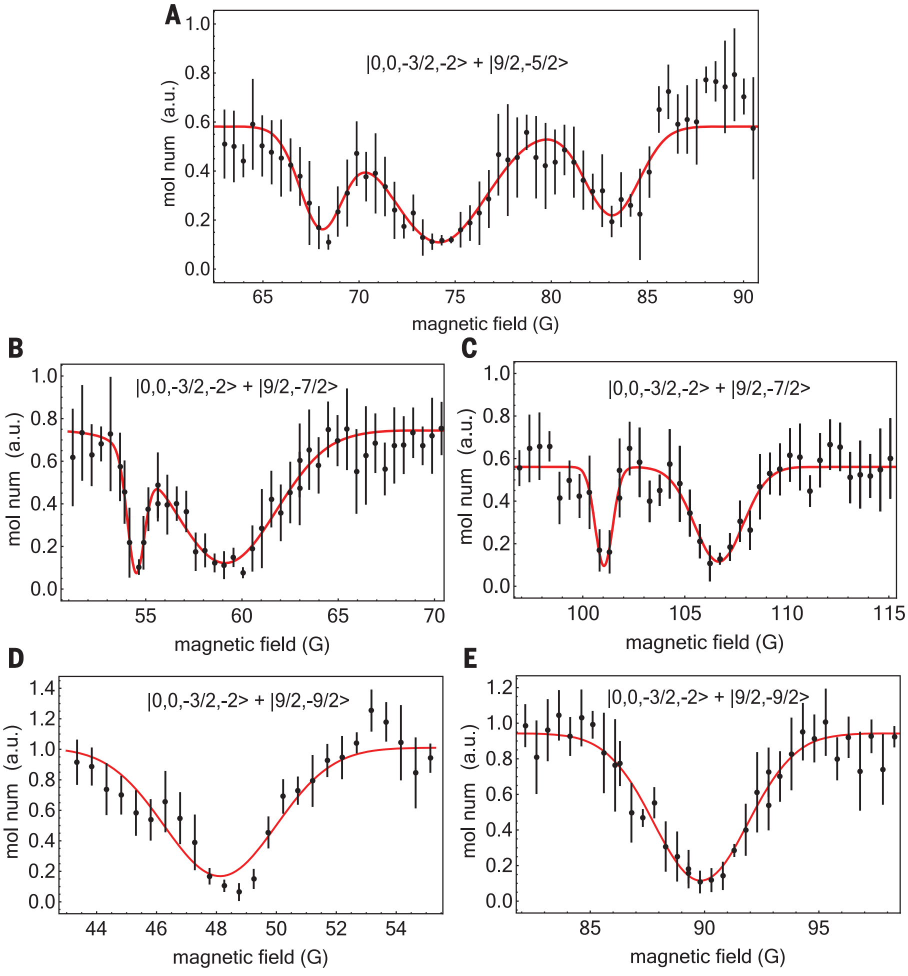

Fig. 4. The resonantly enhanced loss features observed by sweeping the magnetic field. (**A** to **E**) The remaining molecule numbers are plotted as a function of the magnetic field. The loss features provide clear evidence of the atom-molecule Feshbach resonances. The resonance between $|0, 0, -3/2, -2\rangle$ and $|9/2, -7/2\rangle$ at about 101 G is also observed using this method. The solid red lines are phenomenological Gaussian fits with reduced chi-square values of 1.2 (A), 0.81 (B), 0.66 (C), 1.4 (D), and 0.56 (E). The fits are not weighted to error bars. Each data point represents the average of five to eight measurements, and the error bars represent 1 SD of the molecule number.

the exciting possibility of studying resonantly interacting atom-molecule mixtures and may allow creation of ultracold triatomic molecules using magnetic association (*2*).

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ACKNOWLEDGMENTS

Funding: This work was supported by the National Key R&D Program of China (under grant no. 2018YFA0306502), the National Natural Science Foundation of China (under grant no.11521063), the Chinese Academy of Sciences, and the Anhui Initiative in Quantum Information Technologies. **Author contributions:** B.Z. and J.-W.P. conceived the experiment. H.Y., D.-C.Z., and L.L. carried out the experiments. All authors contributed to the analysis of the data and to the writing of the manuscript. B.Z. and J.-W.P. supervised the work. **Competing interests:** The authors declare no competing interests. **Data and materials availability:** All data are available in the main text or the supplementary materials.

SUPPLEMENTARY MATERIALS

www.sciencemag.org/content/363/6424/261/suppl/DC1
Materials and Methods
Figs. S1 to S5
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Data S1

20 June 2018; resubmitted 27 August 2018
Accepted 28 November 2018
10.1126/science.aau5322

Science

Observation of magnetically tunable Feshbach resonances in ultracold $^{23}\mathrm{Na}^{40}\mathrm{K} + {}^{40}\mathrm{K}$ collisions

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Science **363** (6424), 261-264.
DOI: 10.1126/science.aau5322

Brief get-togethers between NaK and K

Cooling molecules to nanokelvin temperatures places them under the tightest quantum mechanical constraints. Studies in this intriguing regime have been limited to diatomics: Two cold atoms can be lured together into weakly associated Feshbach resonances, which lasers can then shift into a more stable molecular state. Yang *et al.* now report the observation of triatomic Feshbach resonances in ultracold collisions between potassium (K) atoms and sodium potassium (NaK) diatomics. The findings potentially set the stage for the preparation and study of ultracold triatomic molecules.

Science, this issue p. 261

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