Controlled state-to-state atom-exchange reaction in an ultracold atom–dimer mixture

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Ultracold molecules offer remarkable opportunities for the study of chemical reactions close to zero temperature. Although significant progress has been achieved in exploring ultracold bimolecular reactions, the investigations are usually limited to measurements of the overall loss rates of the reactants. Detection of the reaction products will improve our understanding of the reaction mechanism and provide a unique opportunity to study the state-to-state reaction dynamics. Here we report on the direct observation of an exoergic atom-exchange reaction in an ultracold atom–dimer mixture. Both the atom and molecule products are observed and the state-to-state reaction rate coefficient is measured. By changing the magnetic field, the reaction can be switched on or off, and the rate coefficient can be controlled. The observed atom-exchange reaction is an effective spin-exchange interaction between the dimer and the atom and may be exploited to study the Kondo effect with ultracold atoms.

Chemical reaction at ultracold temperatures is a fascinating research field and has attracted great interest in recent years1–4. In the past decade, ultracold Feshbach molecules and ground state molecules have been exploited to study ultracold bimolecular reactions, and significant progress has been achieved5–11. However, in these works, the energy released in the reaction is usually much larger than the trap depth, and the reaction products are all lost from the trap. Consequently, only the loss of the reactants can be observed. This makes the direct observation of the reaction products and the study of state-to-state reaction dynamics12 extremely difficult.

The situation will be different if the energy released in the reaction is smaller than the trap depth. Let us consider an atom-exchange reaction between an atom and a dimer, AB + C → AC + B, where AB and AC are both weakly bound Feshbach molecules, and A, B and C are distinguishable atoms. As the binding energy of Feshbach molecules can be precisely controlled by changing the external magnetic field13, it is possible to tune the energy released in the reaction to be small so that the reaction products can still be trapped. In this case, the quantum states of the reaction products may be detected and characterized, and the state-to-state reaction dynamics may be studied.

Scientific interest in reactive collisions involving the uppermost long-range molecules dates back about 40 years and has revived recently due to the creation of Feshbach molecules14,15. In ref. 14, it is predicted that this kind of reactive collision for long-range molecules may be qualitatively different from collisions involving only normal or vibrationally excited molecules, and the reaction might selectively react through a single channel AB + C → AC + B even if many inelastic collision and reaction channels are energetically allowed. Investigations of such atom-exchange reactions can be conducted only in ultracold gases so far since the near-dissociation molecules can be easily destroyed by thermal collisions.

Earlier experimental studies of this kind of reaction A + B → AB + A were performed in ultracold Cs gases, with A and B being different internal states15. The observation of the atom product was reported, whereas the molecule product AB could not be observed because of the absence of Feshbach resonances and the short lifetime of the AB molecules. Besides that, only the loss rates of the molecule reactants were measured. Another experiment that is related to the exchange reaction was performed with three internal states of 8Li, where the influence of the reaction on the overall loss rates was observed16–18. However, the observation of the reaction products was impossible in this work, as the reaction is strongly suppressed for magnetic fields at which the reaction products can be trapped. Therefore, a definitive detection of this kind of atom-exchange reaction and study of the reaction dynamics still remains elusive.

Experimental scheme
Here we report on the direct observation of the AB + C → AC + B reaction and the study of the reaction dynamics in an ultracold 23Na and 40K mixture, where A is the lowest ground hyperfine state |F, mF⟩Na = |1, 1⟩ of 23Na, and B and C are the |F, mF⟩K = |9/2, −5/2⟩ and |9/2, −3/2⟩ ground hyperfine states of 40K, respectively. The AB and AC molecules are the corresponding NaK Feshbach molecules denoted by |1, 1; 9/2, −5/2⟩ and |1, 1; 9/2, −3/2⟩. Since B and C are different internal states of the same atom, the atom-exchange reaction can be regarded as an effective spin-exchange interaction between a NaK Feshbach molecule and a K atom. In our experiment, we prepare approximately 3.0 × 10^5 Na and 1.6 × 10^5 K atoms at about 500 nK in a crossed-beam dipole trap (see Methods). The measured trap frequencies for K are h × (235, 221, 83.5) Hz and the trap depth is calculated to be about k_B × 5 μK for K, with h and k_B being the Planck and Boltzmann constant, respectively.

We first characterize the Feshbach resonance between the A and B (C) atoms at approximately 138 G (131 G; ref. 18) by measuring the binding energies of the AB (AC) molecules. The results are shown in Fig. 1a and the data are fitted using the universal model15, which gives resonance positions of B_0 = 138.7(120) G

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and 130.637(14) G for the two resonances, respectively (see details in Methods and Supplementary Information). The fitted binding energy curves intersect at approximately 130.24 G. Below this magnetic field, the energy of the AB molecule is higher than that of the AC molecule, and thus the reaction is exoergic. The released energy is the difference between the molecule binding energies (see Fig. 1b) and is distributed between the products according to energy and momentum conservation. Therefore, when the kinetic energy acquired by the reaction products is smaller than _the_ trap depth, the products can be trapped—which corresponds to the window of 129.7–130.2 G. This narrow window may be slightly extended towards smaller magnetic fields, because the atom products can quickly lose their kinetic energy by elastic collisions with background atoms and thus may still be trapped.

**Observation of the atomic reaction product**

To observe the atom-exchange reaction, we first prepare the atom–dimer mixture by employing radiofrequency (rf) association\(^\text{19–22}\). To this end, as shown in Fig. 2a, we prepare the Na and K atoms in the A and C states, respectively, then associate the AB molecules from the A + C mixture by applying a 0.5–1 ms Blackman rf pulse with a frequency close to the \(\{9/2, −3/2\} \rightarrow \{9/2, −5/2\}\) transition (Supplementary Information). After association, the desired atom–dimer mixture AB + C is prepared coexisting with the remaining A atoms. If the reaction takes place, B atoms will appear in the mixture. However, direct absorption imaging cannot distinguish between the B atoms and AB molecules, and thus we employ rf spectroscopy to distinguish between them. The AB molecules are measured by a Blackman rf pulse to prepare the AB + C mixture. After the exchange reaction, the AB molecules and the B atoms are transferred to the \([9/2, −7/2]\) state for detection by rf pulses with different frequencies. The molecule dissociation and atom rf spectra. Clear atom peaks are observed for magnetic fields below 130 G, which implies that the exchange reaction may take place at these fields. The duration of the association pulse is 0.5 ms (1 ms) for magnetic fields above (below) 130 G. The AB molecules are dissociated by a 1 ms square rf pulse applied 500 μs after the association pulse. The atomic rf spectra are measured by a 57 μs rf pulse, applied 500 μs after the association pulse. This short pulse is a π pulse with an efficiency of about 90% if on resonance. The dissociation spectra are fitted with the bound-free Franck–Condon lineshape\(^\text{23}\) and the atomic spectra are fitted with the Gaussian model. Error bars represent ±1 s.d.

**Figure 2** | Preparation and detection scheme and observation of the atom products. **a**, The AB molecules are associated from the A + C mixture by a Blackman rf pulse to prepare the AB + C mixture (A atoms not shown). After the exchange reaction, the AB molecules and the B atoms are transferred to the \([9/2, −7/2]\) state for detection by rf pulses with different frequencies. **b**, Molecule dissociation and atom rf spectra. Clear atom peaks are observed for magnetic fields below 130 G, which implies that the exchange reaction may take place at these fields. The duration of the association pulse is 0.5 ms (1 ms) for magnetic fields above (below) 130 G. The AB molecules are dissociated by a 1 ms square rf pulse applied 500 μs after the association pulse. The atomic rf spectra are measured by a 57 μs rf pulse, applied 500 μs after the association pulse. This short pulse is a π pulse with an efficiency of about 90% if on resonance. The dissociation spectra are fitted with the bound-free Franck–Condon lineshape and the atomic spectra are fitted with the Gaussian model. Error bars represent ±1 s.d.
mechanism. Therefore, an unambiguous demonstration of the reaction requires the observation of the molecule product AC. Besides, the detection of the molecule product is equivalent to the direct observation of a spin flip of the Feshbach molecule due to the spin-exchange interaction.

Detecting the molecule product is much more challenging than detecting the atom product. In our experiment, there are essentially two difficulties. The first difficulty is the short lifetime of the molecule product. The second difficulty is that detection of the AC molecule requires rf dissociation, which works well only when being very close to resonance, as this Feshbach resonance is closed-channel dominated. Therefore, we choose to work between 130.1 G and 130.4 G. However, association of the AB molecules from the A + C mixture is inefficient at these fields. Therefore, we change to a different preparation sequence, as shown in Fig. 3a. We first associate the AB molecules from the A + [9/2, −7/2] mixture, and then apply two successive π pulses to transfer the [9/2, −7/2] atoms to the C state. In this way the AB + C mixture is also prepared. To observe the AC molecule products, we dissociate them into the C state via two π pulses. After the exchange reaction, the AC (AB) molecules and the C (B) atoms are transferred into the [9/2, −1/2] (|−7/2⟩) state by rf pulses with different frequencies. 

**Figure 3 | Preparation and detection scheme and observation of the molecule and atom products.**

**a.** The mixture is initially prepared in the A + [9/2, −7/2] state (A atoms not shown), and then the AB molecules are associated with a 1 ms Blackman pulse, after which the K atoms are transferred into the C state via two π pulses. After the exchange reaction, the AC (AB) molecules and the C (B) atoms are transferred into the [9/2, −1/2] (|−7/2⟩) state by rf pulses with different frequencies. 

**b.** Dissociation spectra for AC molecules at different magnetic fields. The molecule dissociation and atom spectra are both measured with 1 ms Blackman rf pulses here, and are applied 100 μs after the second π pulse. Clear AC molecule signals, well separated from the C atom peak, appear at 130.10 G and 130.21 G. At 130.34 G, no molecule signal is observed, where the grey points denote the background in which no association pulse is applied. The dashed line represents the molecule binding energy at this field.

**c.** Atom rf spectra for B atoms. The rf pulse for atoms is a 57 μs short pulse and is applied 50 μs or 0.5–1 ms after the second π pulse. Atom increase is clearly observed at 130.10 G and 130.21 G for a 0.5–1 ms delay. No atom increase is observed at 130.34 G. Error bars represent ±1 s.d.

**Reaction dynamics**

The observation of the reaction products and the characterization of their quantum states provide unique access to studying the state-to-state reaction dynamics, which can be achieved by measuring the time evolutions of the AB reactant and B product numbers. To this end, we choose to prepare the AB + C mixture with the sequence shown in Fig. 2a. After a controllable delay, we dissociate the AB molecules to measure the decay of the molecule reactant, or apply a π pulse to transfer the B atoms to the [9/2, −7/2] state to measure the increase of the atom product. The measured time evolution of the reactant and the product at 129.90 G is shown in Fig. 4a. It is readily seen that the reaction is very fast, with a timescale of a few hundred microseconds, during which the losses of the atom reactant C and product B are negligible. The losses of molecule reactants AB due to collisions with the surrounding atoms cannot be neglected. The reaction dynamics may be described by

\[
\dot{N}_{AB} = -\gamma_{AB} N_{AB} \\
\dot{N}_B = \beta_{BC} N_{AB} 
\]

where \(N_{AB}\) and \(N_B\) are the number of AB reactant and the number of B product, respectively, and \(\beta_{BC}\) is the mean density of the C product from the background atoms due to the imperfection of the π pulses, we compare the atomic rf spectrum between two delay times. At 130.10 G and 130.21 G the atom increase can be clearly observed for a 0.5–1 ms delay, which confirms that the reaction takes place, whereas at 130.34 G no atom increase is observed for 1 ms delay, which also implies that the reaction may be forbidden. The observation of the molecule product is a 'smoking gun', and thus unambiguously demonstrates that the atom-exchange reaction has been observed.

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number of the AB molecules, $a = ((m_B \bar{\omega})/(4\pi k_B T_K))^{1/2}$, with $\bar{\omega}$ the geometric mean of the trapping frequencies of the K atoms, and $T_K$ is the temperature of the K atoms. Note that due to three-body losses in the association process, the temperature of the K atoms increases to about 650 nK after the rf association. The measured state-to-state reaction rate coefficients are shown in Fig. 4b. The reaction rate coefficient can be magnetically controlled and varies from $1.9(3) \times 10^{-10}$ cm$^3$ s$^{-1}$ to $1.1(3) \times 10^{-9}$ cm$^3$ s$^{-1}$ for magnetic fields from 129.44 G to 130.03 G. The exchange reaction is the dominant molecule loss mechanism. At 130.03 G, the measured ratio between the reaction rate $\beta_nC$ and the total decay rate $\gamma_{AB}$ is 80(14)%. This indicates that the reaction selectively reacts through a single channel, as predicted in refs 4,14. This dominance may be understood by the quantum halo character of the Feshbach molecules.

The reaction observed in our experiment may be understood from the point of view of three-body physics. In the case that the two-body scattering lengths $a_{AB}$ and $a_{AC}$ are both much larger than the range of the van der Waals interactions, the atom–dimer reactive scattering may be quantitatively described by the coupled Skorniakov–Ter-Martirosian (STM) integral equations

$$M(0) = M_{AB} + M_{AC} + M_{ABC}$$

In the STM equations, there is an unknown three-body parameter that must be determined to give a quantitative result. The measured reaction rate coefficients may help to determine the three-body parameter by comparing the experiment and theory. Although a quantitative theoretical result is beyond the scope of the current work, the variation of the reaction rate coefficient as a function of the magnetic field near an overlapping Feshbach resonance has been qualitatively discussed in ref. 26. In the limit of zero temperature and large two-body scattering lengths, under the condition of $a_{AC} \ll a_{AB}$, the reaction rate coefficient is predicted to be proportional to $M(a_{AC}) \times a_{AC}^2/a_{AB}^3$, where $M(a_{AC})$ is a log periodic function in $a_{AC}$ (ref. 26). Although these conditions are not fully satisfied in our experiment, the variation of the reaction rate coefficient versus the magnetic field may be qualitatively understood from this formula. At magnetic fields between 129.44 G and 130.03 G, $a_{AB}$ and $M(a_{AC})$ do not change significantly, while $a_{AC}$ has changed from about 500 $a_0$ to about 900 $a_0$. Therefore the reaction rate coefficient increases as the magnetic field increases.

**Conclusions and outlook**

The atom-exchange reaction is also an effective spin-exchange interaction between the NaK Feshbach molecules and the K atoms. The molecule and atom products can be considered as the result of spin flips of the molecule and atom reactants. We expect that at magnetic fields between 130.24 and 130.64 G the inverse exchange collision $AC + B \rightarrow AB + C$ may also be observed. Such an effective spin-exchange interaction is just the interaction required to explore the Kondo effect with NaK Feshbach molecules proposed recently in ref. 27, where the NaK Feshbach molecules serve as magnetic impurities that have spin-exchange interactions with the immersed Fermi sea. Our work therefore opens up a realistic avenue of quantum simulations of the Kondo effect and the related strongly correlated phenomenon

$$M(0) = M_{AB} + M_{AC} + M_{ABC}$$

Our work represents the observation of a quantum-state selected/resolved single-channel-dominated ultracold reaction. The quantum states, the temperature and the released energy are all independently controllable. One unique feature of this reaction is that not only the reactants are ultracold, but also the products are in the ultracold regime since the released energy can be tuned to be very small. This indicates that, by further lowering the temperature, chemical reactions in quantum degenerate gases might be studied. For example, if the B state is initially occupied with a large Fermi energy, and if the released energy in the reaction is precisely tuned smaller than that Fermi energy, the reaction may be suppressed due to Pauli blocking. This opens up the possibility of the investigation of Pauli-blocked chemistry, which is the fermionic counterpart of...
Bose-enhanced superchemistry. Another many-body effect that may be studied is the reaction between Bose polarons. This requires the Na atoms are cooled to the Bose–Einstein condensate and the K atoms are immersed in the condensate as impurities. In this case, the K atoms strongly interact with the BEC and will form Bose polarons. For large repulsive scattering lengths, the repulsive polaron and the attractive polaron have been observed in experiments, which respectively correspond to the free K atoms and the Feshbach molecules in our case. In a system where two internal states of the K atoms can both strongly interact with the BEC and form Bose polarons, the reaction between a repulsive polaron and an attractive polaron, the counterpart of the reaction between an atom and a dimer demonstrated in our work, might be observed.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

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Author contributions

Y.-A.C., B.Z. and J.-W.P. conceived the experiments. J.R., H.Y., L.L., D.-C.Z. and Y.-X.L. performed the numerical calculations. All authors analysed the data and contributed to the writing of the paper. B.Z. and J.-W.P. supervised the work.

Additional information

Supplementary information is available in the online version of this paper. Reprints and permissions information is available online at www.nature.com/reprints. Publisher’s note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations. Correspondence and requests for materials should be addressed to B.Z. or J.-W.P.

Competing financial interests

The authors declare no competing financial interests.
**Methods**

*Ultracold Bose–Fermi mixture preparation.* We load Na from a Zeeman slower and K from a two-dimensional magneto-optical-trap (MOT) into a two-species dark MOT. The atoms are optically pumped to the $\{2,2\}$ and $\{9/2,9/2\}$ states and are then transferred to an Ioffe–Pritchard cloverleaf magnetic trap, where Na atoms are subject to forced evaporative cooling and K atoms are sympathetically cooled. The atomic mixture is then loaded into a crossed-beam optical dipole trap (wavelength 1.064 nm, beam waist 61 μm and 123 μm for horizontal and vertical beams, respectively) and Na atoms are transferred to the A state before further evaporative cooling. At the end of the optical trap evaporation we adiabatically increase the optical trap power to the desired value to hold the atoms and perform the experiments. The Na atoms are always in the A state, and the K atoms are prepared in different internal states by a 50 ms adiabatic rapid passage rf pulse with nearly unit efficiency. The experiments are performed at magnetic fields of around 130 G. The magnetic field is actively stabilized with a stability of better than 10 mG. We prepare K atoms in the $\{9/2,−1/2\}$ or $\{9/2,−7/2\}$ states at a low field before increasing the magnetic field to the desired value, since these two states have no Feshbach resonances at approximately 130 G. K atoms in the C state are prepared from the $\{9/2,−1/2\}$ state by a π pulse transfer at high field. K atoms in different internal states are measured with a high-field imaging technique, and the measured atom numbers are calibrated by comparing with the $σ^−$-cycling transition of $\{9/2,−9/2\}$.

**Characterization of the Feshbach resonances.** The binding energy of the AB Feshbach molecule is measured by the rf loss or dissociation spectrum. For the rf loss spectrum, we prepare the K atoms in the C state, and apply a 0.5–1 s rf pulse to couple the free atom states to the molecular bound states, where the rf field has a Rabi frequency of $\sim 2π \times 1$ kHz for the free atomic transition. The binding energy is then obtained by fitting the loss spectrum (Supplementary Information) with models in ref. 32. For the dissociation spectrum, we associate the AB molecules from the A + C mixture and then dissociate them into the A + $\{9/2,−7/2\}$ state as in the main text. The binding energy is obtained by fitting the dissociation spectrum using the bound–free Franck–Condon lineshape in refs 22,23.

The binding energy of the AC Feshbach molecule is measured using similar methods. For the rf loss spectrum, we prepare free K atoms in the $\{9/2,−1/2\}$ state, and apply a weak rf pulse to observe the loss spectrum. For the dissociation spectrum, the AC molecules are rf associated from the $\{9/2,−1/2\}$ state. Then the remaining K atoms in the $\{9/2,−1/2\}$ state are transferred to the $\{9/2,1/2\}$ state via a π pulse, leaving the $\{9/2,−1/2\}$ state empty for the molecule dissociation. The AC molecules have to be dissociated in such a way because the dissociation from the AC molecule into the A + B free state is significantly suppressed due to the existence of the bound–bound transition.

We fit the measured binding energies with the universal model

\[ E_b = -\hbar^2 / 2\mu (a - \bar{a})^2 \]

where $\mu = m_p m_A / (m_p + m_A)$ is the reduced mass, $\bar{a} = 51 a_0$ is the mean scattering length, with $a_0$ the Bohr radius, and $a = a_0 [1 - \Delta B / (B - B_0)]$ is the scattering length near the Feshbach resonance, with the background scattering length $a_0$, the resonance position $B_0$ and the width $\Delta B$ as the fitting parameters. For Feshbach resonance between A and B, the fitting yields $a_0 = -853(18)$ $a_0$, $B_0 = 138.71(20)$ G and $\Delta B = -34.60(34)$ G, which are in good agreement with previous work. This is an open-channel-dominated resonance with strength $s_{AB} \gg 1$ (ref. 33). For the Feshbach resonance between A and C, we obtain $a_0 = 126(9)$ $a_0$, $B_0 = 130.63(14)$ G and $\Delta B = 4.0(4)$ G. This resonance tends towards closed-channel dominance with the strength $s_{AC} < 1$ (ref. 33). The binding energy measurement gives a resonance at 130.64 G instead of the 129.4 G determined from the enhanced atom loss measurement. The measured binding energies have also been fitted with the coupled-channel calculations (Supplementary Information).

**Reaction dynamics.** The time evolution of the molecule reactant number $N_{AB}$ and atom product number $N_B$ may be described by equation (1), which has the solution

\[ N_{AB}(t) = N_{AB}(0)e^{-\gamma_B t} \]

\[ N_B(t) = \beta \frac{N_{AB}(0)}{\gamma_B} (1 - e^{-\gamma_B t}) + N_B(0) \]

where $N_B(t)$ describes the atom product number accumulated during the association process of the AB molecules. Therefore, the reaction rate coefficient is given by equation (2) in the main text, where $\Delta N_B = N_B(\infty) - N_B(0)$ is the increased atom product number after the reactions, and the mean density of the atom reactant is $n_r = n_A$, with $\alpha = (\mu m_B^3 / 4\pi \hbar^2 T_A)^{1/2}$. Therefore, the reaction rate may be obtained by measuring the increased ratio between the atom product and the reactant $\Delta N_B / n_r$, the initial molecule reactant number $N_{AB}(0)$, and the decay rate of the AB molecule $\gamma_B$. Note that the reaction is very fast, a fraction of the AB molecules may have been reactively lost during the dissociation process. Thus, the rf dissociation rate also needs to be taken into account to correct the initial molecule number (Supplementary Information). Statistical and systematical uncertainties in the atom number, the molecule number, the molecule loss rate and the temperature have all been included to calculate the uncertainty in the final reaction rate.

**Data availability.** The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

**References**

33. Viel, A. & Simoni, A. Feshbach resonances and weakly bound molecular states of boson–boson and boson–fermion NaK pairs. *Phys. Rev. A* **93**, 042701 (2016).