Observation of resonant dipolar collisions in ultracold $^{23}$Na$^{87}$Rb rotational mixtures

Xin Ye, 1 Junyu He, 1 Junyu Lin, 1 Mingyang Guo, 1 Goulven Quéménéner, 2 and Dajun Wang 1,3

1 Department of Physics, The Chinese University of Hong Kong, Hong Kong, China
2 Université Paris-Saclay, CNRS, Laboratoire Aimé Cotton, 91405, Orsay, France
3 The Chinese University of Hong Kong Shenzhen Research Institute, Shenzhen, China

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We report the investigation on dipolar collisions in rotational state mixtures of ultracold $^{23}$Na$^{87}$Rb molecules. The large resonant dipole-dipole interaction between molecules in rotational states of opposite parities brings about significant modifications to their collisions, even when an electric field is not present. In this work, this effect is revealed by measuring the dramatically enhanced two-body loss rate constants in the mixtures. In addition, the dipolar interaction strength can be tuned by preparing the NaRb mixture in different rotational levels with microwave spectroscopy. When the rotational level combination is not of the lowest energy, contributions from hyperfine changing collisions are also observed. Our measured loss rate constants are in good agreement with a quantum close-coupling calculation.

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Following several decades of efforts, the research on ultracold polar molecules (UPMs) has become one of the most active areas in atomic, molecular and optical physics [1]. Breakthroughs in producing samples of ground-state UPMs have made available more and more molecular species [2][9], which, with their different statistics, chemical reactivities and permanent electric dipole moments (PEDMs), are offering us a glimpse of the their great potentials. As early as 2010, ultracold chemical reactions were observed for the first time in the pioneering $^{40}$K$^{87}$Rb experiment at JILA [10][11]. Very recently, both the final and intermediate products following this reaction have been observed by the Harvard group [12], and revealed selected parities of the final rotational states distribution in a magnetic field [13]. For several UPMs without allowed chemical reaction channels, losses were still observed [14][15] and attributed to the formation and trapping light excitation of two-molecule complex [16][17].

Most of the resources of UPMs stem from their rich internal structures and PEDMs. The PEDM makes it possible for polar molecules to interact via the long-range and anisotropic dipole-dipole interaction (DDI). UPMs with strong DDIs have been proposed as a versatile platform for quantum simulation of strongly correlated many-body problems [18][19] and robust quantum information processing [20][21] which are not accessible with ultracold atoms interacting via short range forces.

As the PEDM exists only along the molecular axis, for polar molecules to interact via DDI, a non-zero effective dipole moment must first be induced in the laboratory frame. To this end, the most straightforward method is to place polar molecules inside a dc electric field which induces effective dipole moment by mixing between rotational levels. With both reactive Krb [10] and non-reactive NaRb UPMs [22] in moderate electric fields, strong effects of the DDI manifested as enhanced losses and have already been observed. However, to induce a large effective dipole moment, a rather high electric field with the exact value determined by the magnitude of the PEDM and the rotational constant is usually necessary. Incorporating such high electric fields into an ultracold system is feasible, as been done recently [23], but is still quite challenging as it requires an appropriate and advanced experimental setup [24]. Another way is using an ac electric field to couple two rotational levels with opposite parities directly. For example, the two successive rotational levels $J = 0$ and $J = 1$ can be coupled by a microwave (MW) field, and strong DDI arises from the transition dipole moments (TDMs) [25][26].

In this work, we investigate strong dipolar collisions in absence of external electric fields. To this end, we prepare the ultracold ground-state molecules as an incoherent mixture of the $J = 0$ and $J = 1$ rotational levels. Due to the energy degeneracy of the dipole-dipole coupled pair states ($J = 0, J = 1$) and ($J = 1, J = 0$), two polar molecules with each prepared in these two rotational levels can interact directly via the so called resonant dipole-dipole interaction [Fig. 1(a)]. In the $^{23}$Na$^{87}$Rb system, the effective dipole moments reach $\mu_0/\sqrt{6} = 1.31$ D for the $|J = 0, m_J = 0\rangle + |J = 1, m_J = \pm 1\rangle$ mixture, and $0.55\mu_0 = 1.76$ D for the $|0, 0\rangle + |1, 0\rangle$ mixture. Here $\mu_0 = 3.2$ D is the PEDM of NaRb in their electronic and vibrational ground state and $m_J$ is the rotational projection quantum number onto the $B$ magnetic field axis, taken as the quantization axis. The resonant DDI has been well studied in ultracold Rydberg systems, for example by observing spin-exchange dynamics between single Rydberg atoms trapped in optical tweezers [27]. Although the DDI between UPMs is not as strong as that between Rydberg atoms, they have a much longer lifetime. It is thus still possible to implement high fidelity quantum gates with UPMs in optical tweezers [21]. In our investigation, the effect of the resonant DDI and its
FIG. 1. Resonant dipole-dipole interaction between polar molecules in rotational levels of opposite parities. (a) The $J = 0$ and $1$ rotational levels are connected by the TDM $\mu = \langle 0|\mu|1 \rangle$. Two molecules, with one prepared in each rotational level, can exchange the excitation resonantly which induces a strong DDI without any external electric fields. Here $\mu$ is the dipole operator and $r$ is the inter-molecular distance. (b) Starting from molecules in the $|0,0\rangle$ level, molecules in $|1,1\rangle$ or $|1,0\rangle$ can be prepared selectively by applying a MW $\pi$-pulse and a 50-50 rotational mixtures can be prepared by a $\pi/2$-pulse \cite{28}. (c) Contrast decay of the MW driven Rabi oscillations for the $|0,0\rangle \leftrightarrow |1,1\rangle$ (top) and the $|0,0\rangle \leftrightarrow |1,0\rangle$ (bottom) transitions. The upper limits of the coherence time are 590 $\mu$s and 360 $\mu$s from the fittings (red curves), respectively. A coherent mixture prepared by a $\pi/2$-pulse will become incoherent after several milliseconds.

state dependence are revealed by measuring the loss rates of different rotational mixtures of $^{23}$Na$^{87}$Rb molecules. Compared with that between molecules prepared in a single internal state \cite{14}, the loss in rotational mixtures is enhanced by about one order of magnitude.

Our experimental system has been discussed in detail before \cite{14} \cite{22}. In brief, the experiment starts from ultracold samples of $^{23}$Na$^{87}$Rb molecules in the $|0,0\rangle$ rotational level of the electronic and vibrational ground state ($X^1S^+\text{,} v = 0$) prepared by association of ultracold $^{23}$Na and $^{87}$Rb atoms to weakly bound Feshbach molecules followed by population transfer with a stimulated Raman adiabatic passage (STIRAP) \cite{6}. The molecular sample is trapped in a cigar-shaped optical dipole trap formed by crossing two 1064.4 nm laser beams. To prepare NaRb samples in different rotational states or their mixtures, we apply microwave pulses of different pulse areas \cite{28}. For the detection of $|0,0\rangle$ molecules, we first apply a reversed STIRAP to transfer the ground-state molecule back to Feshbach molecule. Following the magneto-dissociation, the number of atoms is detected with absorption imaging. For molecules in the $J = 1$ levels, we have to apply an additional MW $\pi$-pulse to transfer them back to the $|0,0\rangle$ level before the detection.

A complication for investigating collisions in UPM mixtures is the multiple loss channels. With the constant temperature assumption, the time evolutions of $N_0$ and $N_1$, the numbers of molecules in $J = 0$ and $J = 1$, are governed by the coupled rate equations

$$\frac{dN_0(t)}{dt} = -A\beta_{00}N_0(t)^2 - A\beta_{01}N_0(t)N_1(t),$$

$$\frac{dN_1(t)}{dt} = -A\beta_{11}N_0(t)^2 - A\beta_{01}N_0(t)N_1(t).$$

Here, $\beta_{01}$, $\beta_{00}$, and $\beta_{11}$ are the loss rate constants between $J = 0$ and $J = 1$, between pure $J = 0$ and between pure $J = 1$ molecules, respectively. $A = (2\pi^2m/4\pi k_B)^{3/2}$ is a constant with $k_B$ the Boltzmann constant, $m$ the mass of the molecule, and $\pi$ the geometric mean of the trap frequencies. To measure $\beta_{01}$, contributions from $\beta_{00}$ and $\beta_{11}$ should be subtracted from the total loss. As the collisions for pure $J = 0$ and pure $J = 1$ samples are non-dipolar, the loss rate constants $\beta_{00}$ and $\beta_{11}$ are much smaller than $\beta_{01}$. In fact, it is this large difference which makes the $\beta_{01}$ measurement possible. In a separate experiment, we have tried to study the loss in the non-dipolar mixture of two hyperfine states of the $J = 0$ level. However, as the contributions from different channels are all comparable, we have not been able to extract the loss rate constant between the two hyperfine states reliably.

In the current investigation, $\beta_{11}$ and $\beta_{00}$ are obtained in separate experiments. For measuring $\beta_{11}$, we start by preparing rotationally excited NaRb samples \cite{28} with a resonant $\pi$-pulse on the $|0,0\rangle \rightarrow |1,0\rangle$ or the $|0,0\rangle \rightarrow |1,1\rangle$ rotational transition [Fig. 1(b)]. The transition frequencies are around 4.2 GHz and the frequency separation between $|1,0\rangle$ and $|1,1\rangle$ is 0.655 MHz \cite{25} in presence of a 335 G magnetic field. We then measure the molecule number versus holding time in the optical trap. We use only short holding time to avoid large temperature increase due to the heating effect. After fitting the data to the two-body loss model [Eq. (2) without the $\beta_{01}$ term], we obtain $\beta_{11}$. We repeat the same measurement for sample temperatures from 250 nK to 1.5 $\mu$K to obtain the temperature dependence of the $\beta_{11}$ values for $|1,0\rangle$ and $|1,1\rangle$ samples. For $\beta_{00}$, the previously measured values are re-used \cite{14}.

Figure 2 summarizes the loss rate constants for $^{23}$Na$^{87}$Rb molecules in the three different levels which, not surprisingly, are very similar in both the magnitude and the temperature dependence. As have discussed in \cite{14}, these non-dipolar collisional losses are dominated by the complex formation process which is insensitive to the rotational levels. In what follows, we simply repre-
sent them with

$$\beta_{11} = \beta_{00} = \beta_0 \times (T/T_0)^b,$$

(3)

where $\beta_0 = 3.4 \times 10^{-10}$ cm$^3$ s$^{-1}$, $T_0 = 0.97$ $\mu$K and $b = -0.38$.

To prepare the rotational mixtures, we apply a resonant $\pi/2$-pulse on the $|0,0\rangle \rightarrow |1,0\rangle$ or the $|0,0\rangle \rightarrow |1,1\rangle$ rotational transition. A short pulse duration of 6 $\mu$s is chosen so that losses during the pulse are negligible. To ensure the mixtures are incoherent, we wait for 3 ms before taking the loss data. As illustrated in Fig. $1$ (c), the upper limits of the coherent time constants are less than 1 ms. An important issue here is the need for detecting molecules in both levels. Since each procedure takes several ms to complete, during the detection time for molecules in one level, molecules in the other level always experiences some additional losses. To avoid this complication, for each holding time we measure populations in the two rotational levels in two successive shots.

As shown in Fig. $3$ (a) and (b), we measure the number of molecules only at 0 ms and 4 ms. We use this short holding time to avoid the strong heating effect accompanying the rapid losses. As the loss rate constant is temperature dependent, heating in the course of the measurement always complicates the data analysis. Within 4 ms, the measured temperature change is less than 20% and can be treated as a constant $11$ $14$ $22$. To extract $\beta_{01}$, we fit these data points to Eqs. (1) and (2) simultaneously with $\beta_{00}$ and $\beta_{11}$ fixed from the sample temperatures by Eq. (3). We have also verified experimentally that the $\beta_{01}$ values obtained from the four-point measurement match with those obtained from more points within 20%.

Figure $3$ (c) shows $\beta_{01}$ for collisions between two rotational mixtures, with black circles for the $|0,0\rangle + |1,0\rangle$, and red squares for the $|0,0\rangle + |1,1\rangle$. Comparing with the non-dipolar cases in Fig. $2$ the loss rate constants are 3 to 5 times higher for $|0,0\rangle + |1,1\rangle$ and 10 to 20 times higher for $|0,0\rangle + |1,0\rangle$. The ordering between the different magnitudes of the rates $\beta_{01}$ and the rate $\beta_{00}$ (or $\beta_{11}$) can be qualitatively explained by the DDI strengths achieved in this study, i.e., the higher the resonant dipole moment reached by the rotational mixture, the higher the loss rates. The 1.76 D effective dipole moment for the $|0,0\rangle + |1,0\rangle$ mixture is larger than the 1.31 D one for the $|0,0\rangle + |1,1\rangle$ mixture, and of course larger than the non-dipolar cases $\beta_{00}$ or $\beta_{11}$, resulting in increasing losses in each cases.

For a quantitative understanding of the observations, we perform a time-independent quantum close-coupling calculation $29$, which has successfully described two-body collisions of pure $|0,0\rangle$ NaRb molecule $14$ $22$. As the nuclear spins couple with the rotation and interact with the magnetic field, inelastic, hyperfine state changing transitions can occur, in addition to the loss caused by the complex formation. The experimentally observed loss should be the result of all these possible contributions. We therefore included the proper hyperfine struct-
ture of the molecules at the experimental magnetic field \( B = 335 \text{ G} \), using as a basis set the projection of the atomic nuclear spins \( m^\text{Na}_l \) and \( m^\text{Rb}_l \) onto the quantization axis. Both \(^{23}\text{Na}\) and \(^{87}\text{Rb}\) atoms have atomic nuclear spins \( I = 3/2 \).

For the \(|0, 0\rangle + |1, 0\rangle\) mixture with \( m^\text{Na}_l = m^\text{Rb}_l = 3/2 \) used in the experiment, the theoretical total loss rate constant as a function of the temperature is presented in Fig. 3(c) as a black dashed line. For the \(|0, 0\rangle + |1, 1\rangle\) mixture also with \( m^\text{Na}_l = m^\text{Rb}_l = 3/2 \), this is presented as a red dashed line. An overall good agreement with the experimental results is found, both in magnitude and temperature dependence, considered the number of curves computed and added at the end [29], and the absence of any adjusting parameters. For the \(|0, 0\rangle + |1, 1\rangle\) mixture, the contribution of the complex formation loss only is presented as an orange dash-dot line and that from inelastic, hyperfine changing is shown by the orange short-dash curve. We can see that taking only into account the complex formation loss is not sufficient to get a proper agreement with the experimental data. While the inelastic contribution is 2 to 3 times smaller than the complex loss, it is necessary to account for it.

The difference in the rate constant magnitudes between the two mixtures can be explained by the quantum calculation, as it takes into account the proper specific, complicated dressing of the hyperfine eigenstates in the magnetic field and the strong interplay of the collisional channels via the DDI. This can be seen in Fig. 4 where the adiabatic energies, which act as effective potentials for the entrance collisional channels indicated by arrows, are presented. For the \(|0, 0\rangle + |1, 0\rangle\) mixture, the entrance scattering channel is the lowest one. Therefore it is only affected by other scattering channels above. The channel is mainly governed by an attractive DDI and repulsive centrifugal terms, giving rise to the usual centrifugal barriers as the partial wave quantum number \( l \) increases. This can be clearly seen in the bottom panel of Fig. 4 for \( l = 0, 2 \) and for \( l = 1, 3 \) of the projection quantum number \( m_l \). Even and odd values of \( l \) belong to different symmetries [29]. The barrierless, attractive \( s \)-wave curve (\( l = 0 \)) provides the main and large contribution to the rate constant, reported in Fig. 3(c).

In contrast, for the \(|0, 0\rangle + |1, 1\rangle\) mixture, the entrance scattering channel is higher in energy and lies between many other scattering channels of different hyperfine eigenstates. Because of the strong interplay between the DDI and the hyperfine couplings, the surrounding channels modify the entrance channel of this mixture significantly. As can be seen in the top panel of Fig. 4, the \( s \)-wave curve becomes somewhat pushed up from the channels below, while the \( p \)-wave curve (\( l = 1 \)) becomes somewhat pushed down from the channels above and more attractive. Therefore, the \( s \)-wave curve contribute much less in this case and the \( p \)-wave curve contributes the most instead. The overall rate constant is smaller for this mixture as the \( p \)-wave attractive curve is still less attractive than the \( s \)-wave one for the other mixture. This shows that, apart from the lowest scattering channel, collisions are highly specific to the chosen hyperfine eigenstates of the mixture, i.e., it is possible to further control the resonant DDI by selecting different hyperfine states.

In conclusion, we observe resonant dipole-dipole collisions in incoherent rotational mixtures of ultracold \(^{23}\text{Na}\)\(^{87}\text{Rb}\) molecules. While this resonant DDI is promising for applications in quantum information processing, the proper dressing of the hyperfine eigenstates in a magnetic field as well as their strong interplay with the DDI should be carefully taken into account. Especially, when rotational pairs other than the lowest lying one are used, hyperfine changing can still occur even between single molecules trapped in optical tweezers. This may introduce unwanted leakage which will limit the operation time and reduce the fidelity of quantum gates. A possible remedy for this issue is by applying a small dc electric field to decouple the hyperfine levels. This idea has been successfully adopted to extend the coherent time in microwave coupled rotational levels [30, 31].

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∗ Current address: 5. Physikalisches Institut and Center for Integrated Quantum Science and Technology, University Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany
† goulven.quemener@universite-paris-saclay.fr
‡ djwang@cuhk.edu.hk

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