Microwave coherent control of ultracold polar molecules with rotational-state double resonances

Ting Gong,1, 2 Zhonghua Ji,1, 2 Yuting Liu,1, 2 Yanting Zhao,1, 2 Liantuan Xiao,1, 2 and Suotang Jia1, 2

1 State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan, 030006, China
2 Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi, 030006, China
(Dated: November 19, 2019)

We report an experimental demonstration of microwave-assisted coherent control of ultracold 85Rb133Cs molecules in a ladder-type configuration of rotational-state double resonances, following an observation of Rabi oscillations of the related neighbouring rotational states. The population of middle rotational states, which is coupled with a probe microwave (MW) field, can be reduced by a control MW field. Broadening of spectral splitting and shift of central frequency are observed and both are attributed to the control MW field. Applying Akaike’s information criterion, we conclude that our observed coherent spectra happen near the crossover range of electromagnetically induced transparency and Aulter-Townes splitting. Our work is a significant development in microwave-assisted quantum control of ultracold polar molecules with multilevel configuration, and also offers a great potential in quantum information based on ultracold molecules.

Coherent control of a three-level quantum system with external electromagnetic fields has promoted many intriguing quantum optical phenomena and applications, such as the widely-studied electromagnetically induced transparency (EIT) [1-5] and Aulter-Townes splitting (ATS) [4] effects in atomic system. Based on quantum interference characteristic, EIT has witnessed numerous important applications, including lasing without inversion [5], high-precision magnetometry [6], slow light propagation [7], light storage [8] and quantum transistor [9]. Meanwhile, besides traditional spectral precision measurements including dipole transition moment [10] and lifetimes of coupled levels [11], ATS has also some possible applications due to its coherence, such as recently proposed broadband quantum memory [12].

Along with investigations on atoms, coherent control of molecules increasingly attracts more attentions due to their abundant vibrational and rotational states and easy-adjustable dipole-dipole interaction (which exists only in polar molecules). EIT or ATS effects have been demonstrated in different conditions, including heat pipe [13, 14], hollow-core photonic gap-fiber [15] and photonic microcell [16]. However, Doppler broadening, weak transition dipole moment of nonpolar molecules, or large decay rate of excited electronic state (open system) will limit precise controls and applications of multilevel molecular system. Ultracold polar molecules based on laser cooling provide an ideal platform to eliminate Doppler broadening and to enhance transition dipole moment. In addition, benefiting from the existence of a permanent electric dipole moment, microwave (MW) transition with higher accuracy, resolution and stability, is allowed between rotational levels of polar molecules [17]. Comparing open optical transition, MW field can couple nearly closed rotational levels of the lowest ground state molecules.

MW-assisted coherence also plays important roles in ultracold polar molecules, like simulating quantum magnetism [18], acting as qubits [19, 20], controlling ultracold collision [21, 22] and enhancing evaporative cooling [23]. Nowadays there have been several reports on MW-assisted coherent control in ultracold polar molecules formed via Feshbach resonance [24, 25] and direct laser cooling [26]. However, in all of these experiments, coherent transfer is implemented only between two neighboring rotational states. To our knowledge, the experimental report is still lacking on realizing coherent control of multi-quantum states in ultracold molecules, which is not only significant for fundamental physics of quantum optics in this kind of system, but also essential for some applications, including quantum magnetism [18], topological phase [30] and recently proposed synthetic dimension [31]. In particular, as we know, there are only very few theoretical studies done on the EIT of polar molecules [32, 33], thus experimental realization on this topic is desirable.

In this Letter, we present coherent control in a ladder-type ultracold 85Rb133Cs molecular system with two resonant MW fields, following coherent controls of two-level system with one resonant MW field. The initially populated rotational quantum state, $X^1 \Sigma^+ (v=0, J=1)$ ($v$ and $J$ are vibrational and rotational quantum number respectively, are omitted below) is addressed by short-range photoassociation (PA). A weak probe MW field couples initial state with a middle rotational state ($J=2$) and a strong control MW field couples the middle state with another upper rotational state ($J=3$). We show that the population of $X(0,2)$ state, coupled by the probe MW field, can be reduced in the presence of the resonant control MW field. Broadening of spectral splitting and shift of central frequency are observed and both are attributed to the control MW field. The observed coherent spectra
are found to happen near the crossover range of EIT and ATS by applying Akaike’s information criterion (AIC).

Most details of our apparatus have been reported in our previous papers, such as Ref. [34], in which rotational structures of the lowest ground state of $^{85}\text{Rb}^{133}\text{Cs}$ molecule have been precisely measured. The precooled atomic samples and detection of produced molecules remain the same, and the differences only happen in intermediate excited PA state and time sequence.

Figure 1(a) shows the optical passages of detection and production of RbCs molecules. We use the well-known expression $N = A \cdot \cos(\Omega_t t) \exp(-\gamma_2 t) + C$ to fit these experimental data. Here $A$ and $C$ are amplitude and offset respectively and are both set to be 0.5 according to an ideal two-level interaction model. The second procedure is coherent control of rotational quantum states, the Rabi frequency of MW1 and MW2 fields are 0.42(1) MHz and 0.15(3) MHz respectively. It usually takes lots of time, even impossible, to plot such graphs as in Fig. 2 when MW power is too weak. Thus we use the relationship $\Omega \propto \sqrt{P}$ to obtain an expected Rabi frequency by changing MW power $P$.

According to Refs. [37, 38], in order to observe coherent control of a three-level interaction system, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field. Hence in the following context, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field. Hence in the following context, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field. Hence in the following context, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field. Hence in the following context, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field. Hence in the following context, the Rabi frequency of transition coupled by probe field is expected to be much smaller than that coupled by control field.
FIG. 2. Rabi oscillations of X(0,1)-X(0,2) states with probe field (a) and of X(0,2)-X(0,3) states with control field (b). RbCs molecules are initially populated in the X(0,1) state. It is noticed that in (b) we need to firstly transfer molecule to J=2 using a π pulse of MW1 and then change the irradiated time of MW2.

δ_p. The fitted central frequency is 1988.604 (4) MHz, which is resonant transition of J=1 and J=2 states. We set this value to be the zero point for all of the horizontal coordinates in Fig. 3(a). In the presence of MW2 field, the population of J=2 can be reduced or even absolutely suppressed if MW2 power is high enough, showing a typical quantum coherence phenomenon in three-level system.

As far as early in 1995, Gea-Banacloch et al. [39] had theoretically studied a three-level atomic model for both ladder-type and lambda-type in detail. In 2009, Zhou et al. [32] theoretically studied the case of lambda-type and ladder-type hyperfine sublevels in 85Rb133Cs polar molecule. Referencing the obtained complex susceptibility shown in Eq. 22 in Ref. [32], we can derive expression for ladder-type polar molecule as \( \chi = K(\delta_p + \delta_c + i\gamma_{13})/((\gamma_{12} - i\delta_p)(\gamma_{13} - i\delta_p - i\delta_c) + \Omega_{ce}^2/4) \) . Here \( \delta_c \) is control field detuning, \( \Omega_{ce} \) is effective Rabi frequency of control field and can be written as \( \Omega_{ce} = 2\Omega_cJ_1(Z_{32}^c)/Z_{32}^c \), where \( J_1(Z_{32}^c) \) is the first Bessel function of integer order 1 and \( Z_{32}^c = (\mu_{33} - \mu_{22})E_c/\omega_c \). There \( \mu_{33} \) and \( \mu_{22} \) are permanent dipole moments of J=3 and J=2 states respectively. The rotational dependence of the dipole moment can be written in the form \( \mu_{3,1} = \mu_0 + J(J+1)D_0 \), where \( \mu_0 = 1.215 \) D and \( D_0 \) is estimated to be \( 2.31 \times 10^{-7} \) D [40]. MW amplitude \( E_c \) can be derived from Rabi frequency of control field \( \Omega_c \), by \( E_c = \Omega_ch/\mu_{32} \), here \( h \) is reduced Planck constant and transition dipole moment between J=2 and 3 rotational states \( \mu_{32} \) is calculated to be \( \sqrt{9/35}\mu_0 \) by simplifying it to be the value between \( J = 2, m = 0 > and \( |J = 2, m = 0 > \) sublevels based on angular momentum coupling rule and 3-j symbol, where m represents J projection on to the axis defined by the microwave field [11]. Thus \( \Omega_{ce} \) is dependent on \( \Omega_c \) and decreases gradually with an oscillation from the initial value (like Fig. 2(a) in Ref. [32]). Substituting all parameters, we find that \( \Omega_{ce} \rightarrow \Omega_c \) is valid as long as \( \Omega_c \) is lower than \( 10^2\gamma_{12} \) with the maximum deviation of 0.02% at \( 10^5\gamma_{12} \).

It is known that imaginary part of the susceptibility expression leads to absorption characteristic of probe field and also population variety of molecule states coupled by this probe field. By setting \( \gamma_{12} \), \( \gamma_{13} \), \( \Omega_{ce} \), \( \delta_c \) and \( K \) as parameters, the fitting curves are shown with green lines in Fig. 3(a). \( K \) is only a coefficient influencing the spectral amplitude. For all graphs, \( \gamma_{12} \) and \( \gamma_{13} \) are expected and truly verified to be unchanged, with averaged fitting values of 0.2(1) MHz and 0.04(4) MHz respectively. It is reasonable to find that the fitting value of \( \gamma_{12} \) agrees with the value derived from Rabi oscillation in Fig. 2(a). We plot the observed spectrum splitting and fitted \( \Omega_c \) (they are expected to be equal) versus derived Rabi frequency of control field in Fig. 3(b). It is obvious that they are both linear dependent on Rabi frequency of MW2 field. The fitted linear coefficients are theoretically expected to be both 1 in an ideal condition, however they are both experimentally fitted to be 0.72(3). Considering the large errors in photoionization detection, they are roughly agreeable.

FIG. 3(c) shows the variety of central frequency \( \delta_c \). We find there is a frequency shift and attribute it to be the well-known a.c. Stark effect or light shift. In textbooks, like Ref. [42], the eigenvalues of two levels, coupled with an electromagnetic field with frequency detuning \( \delta \) and Rabi frequency \( \Omega_c \), are calculated to be \( \Delta_\pm \approx \delta/2 - (\delta^2 + \Omega^2)^{1/2}/2 \). To explain our observed central frequency shift, we focus on \( J=2 \) state, which is mainly influenced by strong control MW2 and detected by weak probe MW1. The energy level variety can be written as \( \Delta_\pm \approx \delta/2 - (\delta^2 + \Omega^2)^{1/2}/2 \). We use this expression to fit experimental data, resulting \( \delta \) to be 0.71(6) MHz. We need to notice that the above fitting is based on an ideal two-level system. In fact there are \( (2J+1)(2I_{Rb} + 1)(2I_{Cs} + 1) \) hyperfine sublevels in 85Rb133Cs polar molecule, where nuclear angular momenta \( I_{Rb}=5/2 \) for 85Rb and \( I_{Cs}=7/2 \) for 133Cs. According to the Refs. [40, 43], the hyperfine sublevels of \( J=2 \) and \( J=3 \) rotational states cover ~MHz for 85Rb133Cs molecule. The total distributions and transition probabilities determine the finally observed a.c. Stark shift.

So far, we have used an universal formula to analyze our observed coherence spectra and don’t attribute these spectra into EIT or ATS, which are two extremes of three-level coherent spectroscopy. Recently, investigating on discernment between EIT and ATS has become an active topic [37,38,44-49]. Among these work, AIC is proposed to discriminate EIT and ATS from an experimental
FIG. 3. Coherent control of ultracold polar molecules with ladder-type rotational states. (a) The probe detuning-dependent population of $J=2$ rotational state in the absence and presence of control field. The green, purple and red curves in each spectrum are the fitting results based on universal, EIT and ATS formulas respectively. The Rabi frequency of control field are labeled in each spectrum. Based on the universal formula, the splitting of two peaks and the fitted central frequency are displayed in (b) and (c) respectively.

viewpoint [37]. It has been employed widely to quantitatively determine the relative weights of the effects of EIT and ATS in various systems, including cold atoms [44], open V-type hot molecules [45], mechanical oscillators [46], whispering gallery mode microcavities [47], plasmonic waveguide-resonators [48] and superconducting quantum circuits [49]. Here we apply AIC to ultracold polar molecule. The lineshapes of EIT and ATS can be expressed as $A_{EIT} = C_{\pm}^{2} / (\gamma_{+}^{2} + \delta_{0}^{2}) - C_{\pm}^{2} / (\gamma_{-}^{2} + \delta_{0}^{2})$ and $A_{ATS} = C^{2} [1 / (\gamma^{2} + (\delta_{p} - \delta_{0})^{2}) + 1 / (\gamma^{2} + (\delta_{p} + \delta_{0})^{2})]$ respectively [37]. Here $C$ and $C_{\pm}$ stand for the amplitudes of Lorentzian curves, $\gamma$ and $\gamma_{\pm}$ for the linewidths respectively, $\delta_{0}$ for the detunings from resonant frequency. The fitting results are shown with purple and red curves for EIT and ATS models. The per-point AIC contribution of EIT (ATS) model is quantitated by $\omega_{EIT(ATS)} = \exp(-I_{EIT(ATS)}/2N) / [\exp(-I_{EIT}/2N) + \exp(-I_{ATS}/2N)]$, where $N$ is the number of data points and $I = N \cdot \log(\sum_{j=1}^{N} \epsilon_{j}^{2}/N) + 2M$. Here $M$ is the number of fitting parameters and $\epsilon_{j}^{2}$ is the residual of experimental measurement from the fitted model. The weight of EIT model for $\Omega_{c} = 0.29, 0.87$ and $1.39\gamma_{12}$ is 0.50(1), 0.48(1) and 0.47(1) respectively. Knowing that $\omega_{EIT} + \omega_{ATS} = 1$, these weights show that these three coherent spectra all locate near the crossover point (0.5) between EIT and ATS. To separate EIT and ATS more obviously, $\Omega_{c}$ needs to be varied largely and can not be realized under our experimental conditions.

In conclusion, we presented an experimental realization of coherent control of rotational quantum states of ultracold polar molecules when single resonance in two-level and double resonances in three-level are satisfied. In two level case, the Rabi oscillations of neighboring rotational states are observed. In three-level case, the coherence control is performed by monitoring the population of middle rotational quantum state in the presence of control MW field. We confirmed that the fitted spectral splitting from three-level coherence spectrum agrees with the derived value from two-level Rabi oscillation. The central frequency shift is attributed to the a.c. Stark effect induced by the control MW field. By employing AIC to the experimental spectra, it was found that our observed coherent spectra happen near the crossover range of EIT and ATS. To observe ATS-dominated spectrum, it is expected to improve irradiated power of the control field. To observe EIT-dominated spectrum, it is expected to decrease probe field power and ensure enough SNR by improving detection sensitivity and increasing molecule production rate. The present work opens many possibilities for further investigation based on coherence in ultracold molecules. In particular, it makes a significant development in microwave-assisted quantum control of ultracold polar molecules with multilevel configuration, and offers a great potential in quantum information based on ultracold molecules.

We acknowledge Prof. J. M. Hutson from Durham University for his theoretical calculation on the electric dipole moment of rotational states. We thank Dr. Z. H. Li for his early effort on this work. Dr. J. Z. Wu from Zhejiang University for useful discussions. This project was supported by the National Key R&D Program of China (Grant No. 2017YFA0304203), the Na-
