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Microwave-assisted coherent control of ultracold polar molecules in a ladder-type configuration of rotational states

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We demonstrate microwave-assisted coherent control of ultracold ⁸⁵Rb¹³³Cs molecules in a ladder-type configuration of rotational states. Specifically, we use a probe and a control MW field to address the transitions between the $J = 1 \rightarrow 2$ and $J = 2 \rightarrow 3$ rotational states of the $X^{1}\Sigma^{+}(v = 0)$ vibrational level, respectively, and use the control field to modify the response of the probe MW transition by coherently reducing the population of the intermediate J = 2 state. We observe that an increased Rabi frequency of the control field leads to broadening of the probe spectrum splitting and a shift of the central frequency. We apply Akaike's information criterion (AIC) to conclude that the observed coherent spectral response appears across the crossover regime between 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.

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Introduction

The development of coherent control technology based on a three-level quantum system over the last twenty years has shed new light on many intriguing quantum optical phenomena. The most prominent examples include the widely studied electromagnetically induced transparency (EIT)¹⁻³ and Autler-Townes splitting (ATS)⁴ effects. With exploration of quantum interference, EIT effects are central to many applications including lasing without inversion,⁵ high-precision magnetometry,⁶ slow light propagation,⁷ light storage⁸ and quantum transistors.⁹ On the other hand, ATS has important applications within the traditional area of precision measurements such as to measure the dipole moment¹⁰ and lifetime,¹¹ using the associated Stark shift effects, while the coherence associated with ATS has been proposed for broadband quantum memory.¹²

Most of the investigations mentioned above are related to atoms. Compared with atoms, polar molecules hold richer internal levels and possess permanent electric dipole moments. The permanent electric dipole moment is particularly valuable from a quantum control perspective since it enables sensitive external field manipulation and leads to long-range anisotropic many-body interactions. Remarkably, with Doppler broadening suppressed, ultracold polar molecules are promising candidates for quantum many-body physics,^{13,14} precision measurements of fundamental constants¹⁵ and quantum computation.^{16,17}

For all the applications, preparation of ultracold polar molecules is the prerequisite condition for the coherent control of the internal state. Two classes of methods have been developed over the last two decades to produce ultracold molecules. First, direct laser cooling and magneto-optical trapping (MOT) have been applied to realize the production of SrF18 and CaF19,20 molecules. However this method requires nearly closed lasercooling transition which can only be applied to a few molecular species. Another promising approach is based on either magnetoassociation (MA) or photoassociation (PA), which starts from colliding ultracold atoms and then associates them using a magnetic field or a photon. The MA method typically starts from weakly bounded Feshbach molecules in electronic ground states.²¹⁻²⁴ A stimulated Raman adiabatic passage (STIRAP) transfers the population to the ground rovibrational level.^{25,26} On the other hand, molecules formed by PA are usually in highly excited rovibrational states initially, which may continuously decay to lower rovibrational levels via spontaneous processes. Short-range PA is a particularly appealing method because it offers a simple pathway for efficient production of ground state molecules from atoms,²⁷⁻³¹ particularly for species not amenable to MA.³²

We notice that three-level coherent manipulation has been involved in the preparation of ultracold molecules in ground

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states by STIRAP, where coherent spectroscopy is used to determine the resonance frequency between optical transitions.²¹⁻²⁴ After preparation in the ground state, ultracold polar molecules provide an ideal platform to implement precise microwave (MW) coherent control in rotational states,^{17,33–38} which has many applications, such as quantum magnetism,³⁹ topological phase⁴⁰ and synthetic dimensions.⁴¹ However, most of the previous studies of three-level coherent control in the MW range were focused on Rabi oscillation. Few efforts have been made to directly investigate the ATS and EIT spectra for the rovibrational levels. One example is the recent ATS spectroscopy for investigating the dipolar collision of ultracold ²³Na⁴⁰K molecules.³⁸ However these previous studies typically explored the quantum coherent effect in the widely studied lambda-system. Even though, ladder-type quantum coherent spectroscopy attracts growing interest for a variety of applications, such as synthetic dimensions.⁴¹ It is noticed that the research on the control of a three-level ladder system forms the basis for multilevel control of many-level rovibrational states, which may have application in expanding the quantum computation ability, such as encoding quantum information in multiple states as qudits.⁴² We note that recently, Blackmore et al. reported the MW control in a rotational ladder system in ⁸⁷Rb¹³³Cs molecules.⁴³

Each molecule has its own characteristics. ⁸⁵Rb¹³³Cs molecules attract attention benefiting from their special characteristics: sizable permanent electric dipole moment⁴⁴ enables easy alignment for quantum simulation;⁴⁵ avoidable immiscibility of its components, which is different from its isotopic components,⁴⁶ provides possibility to realize molecular Bose–Einstein condensation; their significant inelastic collisional cross-section with co-trapped Cs atom also supports molecule purification in the lowest vibronic state.⁴⁷ Previous work have shown that short-range PA can produce ⁸⁵Rb¹³³Cs molecules in the ground state.³⁰ MW coupling and MW coherent control of ultracold ⁸⁵Rb¹³³Cs molecules have been reported to obtain a rotational constant⁴⁸ by measuring the resonant frequencies and coherence time by observing the associated Rabi oscillations between two rotational levels within the ground vibrational state.⁴⁹

In this work, we present a coherent control of ultracold polar molecules formed by short-range PA in a ladder-type rotational system with probe and control MW fields. The broadening of spectral splitting and shift of central frequency are observed to be dependent on the Rabi frequency of the control MW field. We investigate the coherent spectroscopic response of Rabi frequency of the control field and discriminate EIT and ATS regimes by applying the Akaike's information criterion (AIC).⁵⁰

Experimental setup

Fig. 1(a) shows the optical transitions we used to produce and detect ${}^{85}\text{Rb}{}^{133}\text{Cs}$ molecules. The intermediate level was chosen as $2{}^{3}\Pi_{0-}(\nu = 11, J = 0)$ for that this state is theoretically expected to have only J = 1 distribution for $\nu = 0$ vibrational state³⁰ and has been verified in our measurement. Starting from the simple distribution, we use two coherent MW fields, probe and control fields, to drive the three-level control as shown in Fig. 1(b). Here



Fig. 1 (a) Optical transitions for production and detection of ⁸⁵Rb¹³³Cs molecules. (b) A ladder-type configuration of rotational states for MW coherent control. (c) Timing for the molecular sample preparation, coherent control and measurement sequences in this work.

each of the two fields is generated by using a MW signal generator (Stanford Research Systems, SG386) followed by an power amplifier (Mini-Circuits, ZHL-4W-422+), before feeding to the experiment with a homemade microwave antenna. The two signal generators share a common radio frequency clock to ensure the long-term phase stability. Two fast switches (ZFSWA2-63DR+) are used to switch on/off these MW pulses in less than 35 ns. We measure the radiant power using a microwave power meter (NRP-Z51, R&S) with a circle probe of radius 1.15 cm.

The timing sequence shown in Fig. 1(c) can be divided into three stages: molecule production, coherent control, and statedependent molecule detection. The experiment starts with 1×10^{7} ⁸⁵Rb atoms in the 5S_{1/2} (F = 2) state with a density of $8 \times 10^{10} \text{ cm}^{-3}$ and $2 \times 10^{7} \text{ }^{133}\text{Cs}$ atoms in the $6S_{1/2}$ (F = 3) state with a density of 1 \times 10¹¹ cm⁻³. An intense PA laser from a Ti:sapphire laser is focused onto the overlapping atomic samples, with a total power of 1 W and a beam waist of about 150 µm for 96 ms. With the PA laser frequency tuned to 11803.86 cm⁻¹, ultracold ⁸⁵Rb¹³³Cs in the X¹ $\Sigma^+(\nu = 0, J = 1)$ ground states can be formed. The production rate of ⁸⁵Rb¹³³Cs molecules is around 1×10^4 molecules per second. The temperature of the formed molecules is around 100 µK. Next, the ⁸⁵Rb¹³³Cs molecules are coherently controlled by the two MW fields, which will be detailed later. Finally, the statedependent population detection is realized by depletion spectroscopy⁵¹ and photoionization techniques. For this purpose, a diode laser, with an intensity of 8 mW and a beam waist of 1 mm, is introduced to deplete the population of the J = 2state. Here the depletion laser frequency is locked at 466.8575 THz, which is resonant with transitions from $X^{1}\Sigma^{+}(\nu = 0, J = 2)$ to $2^{3}\Pi_{0+}$ (v = 8, J = 1)⁵² Once the depletion laser is on the resonant transition, the intensity of molecules from v = 0 vibrational state decreases. After that, a photoionization (PI) laser, a pulse duration of 7 ns and a pulse energy of 2 mJ, is applied to ionize ⁸⁵Rb¹³³Cs molecules in the $X^{1}\Sigma^{+}(\nu = 0)$ vibrational state. Finally, an electric field accelerates these photoionized ions to a pair of microchannel plates. The fractional depletion indicates the relative population of the rotational states of interest. The whole experimental sequence is repeated at a 10 Hz rate.

Experimental results and analysis

To implement coherent control on a three-level system, we need to know Rabi frequencies coupled by the probe and control fields and the coherence decay rate of the associated transitions. The Rabi frequency of probe field $\varOmega_{\rm p}$ and the coherence decay rate γ_{12} of J = 1 and J = 2 are derived from the Rabi oscillation signals driven by the probe field alone.⁴⁹ To observe Rabi oscillation between J = 2 and J = 3 states, we first irradiate the molecules in the initial I = 1 state with a π pulse using the probe MW (f = 1988.61 MHz) so as to transfer the molecules to the I = 2 state. We then turn on the control MW for a variable duration to observe the Rabi oscillation by recording the population of molecules in the I = 2 state. Fig. 2 shows our observed Rabi oscillation between J = 2 and J = 3 rotational states. In this measurement, the frequency of control MW is resonant with the transition between I = 2 and I = 3 states (2982.82 MHz).⁴⁸ We fit these data with a simple oscillatory decay function as

$$N = A\cos(\Omega_{c}t)\exp(-\gamma_{23}t) + C.$$
 (1)

With the fit, we retrieve the Rabi frequency for the control field $\Omega_c = 0.42(1)$ MHz, and the coherence decay rate $\gamma_{23} = 0.15(3)$ MHz, respectively, where *A* and *C* are the oscillation amplitude and offset, respectively.

The measured value of Ω_c allows us to derive the transition dipole moment (TDM) between J = 2 and 3 states as $\mu_{23} = \Omega_c \hbar/E$, where \hbar is the reduced Planck constant. More specifically, the MW amplitude E is related to power P through the intensity as $I = 1/2c\varepsilon_0 E^2 = P/\pi r^2$. We therefore obtain $\mu_{23} = \sqrt{(c\varepsilon_0 \pi r^2 \hbar^2)/2P} \cdot \Omega_c = 0.59(4)$ debye, where r is the radius of the MW power meter, c is the speed of light, and ε_0 is the permittivity in vacuum. By taking into account the transmission loss of about 94(5)% through a quartz window, we estimate power P to be 0.282(15) mW as measured by a power meter. To compare with the calculated value, we utilize the same operation as dealing



Fig. 2 Rabi oscillation between $X^{1}\Sigma^{+}(v = 0, J = 2)$ and $X^{1}\Sigma^{+}(v = 0, J = 3)$ states under a control MW field. Ultracold ⁸⁵Rb¹³³Cs molecules are initially populated in the J = 1 state by spontaneous decay, then transferred to J = 2 by a π pulse of probe MW, followed by irradiation of control MW. Every experimental point is an average of 64 times, and the error bars indicate the standard deviations.

with TDM μ_{12} between J = 1 and 2 states in ref. 49, but here for J = 2 and 3 states μ_{23} is expected to be $\mu_{23} = \sqrt{(9/35)}\mu_0$,⁵³ by simplifying it to be the value between $|J = 2, M = 0\rangle$ and $|J = 3, M = 0\rangle$ under a plane-polarized light with the electric vector along the *z* position,^{49,54} where μ_0 is adopted to be 1.225(11) debye³⁵ by considering that μ_0 for ⁸⁵Rb¹³³Cs is only about 9 parts in 10⁷ smaller than for ⁸⁷Rb¹³³Cs.⁴⁹ Thus our measured value (0.59(4) debye) shows an agreement with the calculated value (0.62 debye) under the uncertainty of error.

To clearly observe the quantum interference of the threelevel system, it is best to have the probe Rabi frequency $\Omega_{\rm p}$ to be smaller than the relaxation rate γ_{12} ,^{50,55} which is approximately 0.67 MHz by the *in situ* measurements.⁴⁹ On the other hand, a too low $\Omega_{\rm p}$ leads to a weak signal and a poor signal-to-noise ratio (SNR), even with many rounds of measurements as those in Fig. 2. We adjust Ω according to $\Omega \propto \sqrt{P}$, and choose $\Omega_{\rm p} \sim 17$ kHz in the following measurements as a compromise between the preference of a weak field and decent SNR.

Fig. 3 plots the J = 2 rotational population excited by the probe field under different Rabi frequencies of the control MW field. In the absence of the control MW, displayed in Fig. 3(a), the spectrum shows an expected Lorentzian lineshape as a function of probe field detuning δ_p , with a linewidth around 0.13(4) MHz, arising from the unresolved hyperfine structure.⁴⁹ The fitted central frequency is 1988.604(4) MHz, that is the resonant transition frequency of J = 1 and J = 2 states. We set this value to be the zero point for all the horizontal coordinates in Fig. 3. In the presence of control MW shown in Fig. 3(b–e), the population of J = 2 is reduced when the control MW power increases. The reduction of J = 2 population is associated with the reduced absorption of the probe field. Therefore, Fig. 3(b–e) shall be interpreted as a control field induced transparency to the probe transmission.

The linear susceptibility of the three-level system has been derived in different contexts. A sharp difference between the atomic system⁵⁶ and here is the existence of a permanent dipole moment (PDM) in the polar molecules. The three-level dynamics in the presence of PDM has been studied by Zhou *et al.*⁵⁷ They found that multi-photon transition case with virtual level(s) can happen in this kind of system, and an oscillatory feature will appear in the three-level transition process if two PDMs of molecular states coupled by the control field are comparable. Here we combine the final susceptibility (eqn (22) of ref. 57) with the density-matrix element of the three-level system (eqn (3) in ref. 56), and infer that the three-level susceptibility of polar molecules in a ladder configuration should be given as,

$$\chi = \frac{K(\delta_{\rm p} + \delta_{\rm c} + i\gamma_{13})}{(\gamma_{12} - i\delta_{\rm p})(\gamma_{13} - i\delta_{\rm p} - i\delta_{\rm c}) + \Omega_{\rm ce}^2/4}.$$
 (2)

In this general expression, we define $\Omega_{ce} = 2\Omega_c J_1(Z_{32}^c)/Z_{32}^c$ as the effective Rabi frequency related to the control field, with *K* set as unity for simplicity of numerical analysis. $J_1(Z_{32}^c)$ is the first order Bessel function for the variable $Z_{32}^c = (\mu_3 - \mu_2)E_c/\omega_c$. μ_3 and μ_2 are respectively PDMs of states coupled by the control



Fig. 3 Coherent spectra of ultracold ⁸⁵Rb¹³³Cs molecules in ladder-type rotational states. The population of J = 2 is measured in the absence (a) and presence (b–e) of the control field with the detuning of the probe field. The curve in (a) is a Lorentz fitting, and the red solid lines in (b–e) spectra are the fitting results based on the imaginary part of eqn (2) under approximation of $\Omega_{ce} \sim \Omega_{c}$. The green short dash dotted line and blue dash line are the fitting lines using the EIT model and ATS model, respectively. The probe field and control field are turned on for 1 ms simultaneously (see Fig. 1(c)). The Rabi frequency of the probe field is fixed as 17 kHz, while the Rabi frequencies of the control field are 0, 0.055, 0.09, 0.26, and 0.42 MHz for (a–e) respectively. Every experimental point is an average of 64 times, the error bars indicate the standard deviations.

field, while μ_{23} is their TDM. E_c is the amplitude of the control field. As mentioned before, if μ_3 is comparable with μ_2 , the coherent phenomenon will be complicated even for three-level transition because of an oscillatory feature in the Bessel function as a function of E_c .⁵⁷ Fortunately, here the difference between μ_3 and μ_2 is only $6D_{\mu_0} = 1.4 \times 10^{-6}$ debye,⁴⁹ and therefore we have $\Omega_{ce} \approx \Omega_c$ under the typical experimental condition of moderate MW power.

It is known that the imaginary part of susceptibility leads to the absorption of the probe field, and corresponds to state population variation induced by the couple field. By setting γ_{12} , γ_{13} , Ω_c , δ_c and *K* as parameters, the fitting results are shown with red solid lines in Fig. 3(b–e). For all the graphs, γ_{12} and γ_{13} are expected and truly verified to be unchanged, with averaged values of 0.2(1) and 0.02(1) MHz, respectively. The large uncertainty of fitted γ_{12} obtained here and γ_{12} derived from Rabi oscillation is attributed to the fluctuation of atom density, stability of photoionization laser, detection sensitivity of molecules, *etc.* in a large span of experiment running time.

We in addition observe a shift of transition frequency between J = 1 and 2, δ_c , induced by the control MW field. The

results are presented in Fig. 4(a). We attribute the shift to the A.C. Stark effect or the light shift. Since the J = 2 state is mainly influenced by the strong control MW field while the perturbation by the weak probe is negligible, we shall write down the total shift of the effective two-level system as:⁵⁸

$$\delta_{\rm c} = \frac{\delta_{\rm c0}}{2} - \frac{\sqrt{\delta_{\rm c0}^2 + \Omega_{\rm c}^2}}{2}.$$
 (3)

The shift is divided into two parts: the initial detuning and the induced shift by Ω_c . The initial detuning of control MW δ_{c0} is fitted to be 710(60) kHz. The factor influencing this detuning arises mainly from the unresolved hyperfine structure. The molecules in rotational state *J* have $(2J + 1)(2I_{Rb} + 1)(2I_{Cs} + 1)$ hyperfine sublevels, where $I_{Rb} = 5/2$ and $I_{Cs} = 7/2$ are the nuclear spins for ⁸⁵Rb and ¹³³Cs, respectively. According to ref. 59, the dominant hyperfine interactions in states with J > 0 are nuclear electric quadrupole interactions. The allowed microwave lines for $J = 2 \rightarrow J = 3$ in the absence of a control field are expected to be spread over about 567 kHz in the absence of a magnetic field,⁶⁰ which is comparable with δ_{c0} .

As in Fig. 3(b–e), we observe an increased splitting of the spectrum, $\Delta_{\rm sp}$, as a function of the control field Rabi frequency $\Omega_{\rm c}$. This observation is similar to those in atomic systems where one finds the relationship $\Delta_{\rm sp} = \alpha \sqrt{\Omega_{\rm c}^2 + \delta_{\rm c}^2} = \alpha \Omega_{\rm c}'$. Here $\Omega_{\rm c}'$ is the generalized Rabi frequency of the control MW field and α is a fitting parameter. We plot the generalized Rabi frequency-dependent curve of the observed spectrum splitting in Fig. 4(b) and the α is extracted to be 0.62(2). The deviation from unity is attributed to the error of $\Omega_{\rm c}$ inferred from the relationship $\Omega \propto \sqrt{P}$.



Fig. 4 Control MW field-induced central frequency shift (a) and spectrum splitting (b). The experimental data in (a) are obtained by subtracting the fitting central frequencies in MW coherent spectra and the resonance transition frequency between J = 1 and J = 2. The data in (b) are frequency differences of two peaks fitted by Gaussian function. The curve in (a) is the fitting result based on eqn (3), and the line in (b) shows a linear fitting.

The general formula by eqn (2) allows us to analyze the coherent spectrum uniformly. However, it is more instructive to characterize the lineshape of coherent spectroscopy which can be the regime of EIT, ATS, or the cross-regime in between. The characteristic difference between EIT and ATS and its quantification has been an interesting topic to be studied actively in recent years.^{50,55,61–66} Among these works, AIC is proposed to discriminate EIT and ATS from an experimental viewpoint.^{50,61,63–66} It has been employed widely to quantitatively determine the relative weights of the effects of EIT and ATS in various systems, including cold atoms,⁶¹ mechanical oscillators,⁶³ whispering gallery mode microcavities,⁶⁴ plasmonic waveguide-resonators⁶⁵ and superconducting quantum circuits.⁶⁶ Here we apply AIC to our system of ultracold polar molecules.

Specifically, the lineshapes of EIT and ATS can be expressed as $A_{\rm EIT} = C_+/(1 + (\delta - \varepsilon)^2/(\gamma_+^{-2}/4)) - C_-/(1 + \delta^2/(\gamma_-^{-2}/4))$ and $A_{\rm ATS} = C_1/(1 + (\delta + \delta_1)^2/(\gamma_1^{-2}/4)) + C_2/(1 + (\delta - \delta_2)^2/(\gamma_2^{-2}/4))$, respectively.⁶¹ Here C_1 , C_2 and C_{\pm} are the amplitudes of Lorentzian curves, γ_1 , γ_2 and γ_{\pm} are the linewidths, respectively, and ε , δ_1 and δ_2 are the detunings from the resonant frequencies. The fitting results are shown in Fig. 3(b–e) with green and blue curves for EIT and ATS models. The per-point AIC contribution of the EIT (ATS) model is quantitated by $\omega_{\rm EIT(ATS)} = \exp(-I_{\rm EIT(ATS)}/2N)/[\exp(-I_{\rm EIT}/2N) + \exp(-I_{\rm ATS}/2N)]$, where N is the number of data points of each spectrum and

 $I = N \log \left(\sum_{j=1}^{N} \varepsilon_j^2 / N \right) + 2M$. Here *M* is the number of fitting

variables and ε_j^2 is the residual of experimental measurements from the fitted model. The weights of the EIT model in Fig. 3(b–e) are 0.57, 0.50, 0.48, and 0.45 with an error of 0.05 as Ω_c increases, while the weights of ATS are the values subtracted by one. This tendency suggests that the EIT effect dominates the lineshape of the spectrum when Ω_c is small, while ATS dominates the lineshape of the spectrum when Ω_c is large.

Conclusion

In conclusion, we have reported microwave-assisted experimental realization of coherent spectroscopy of ultracold polar molecules with a ladder-type configuration of rotational states, formed by the short-range PA method. The coherent control is achieved by monitoring the population of middle rotational states in the presence of a control MW field. The spectrum splitting is proportional to the Rabi frequency of the control field. The observed frequency shift in the coherent spectra is attributed to the A. C. Stark effect induced by the control MW field, while the initial detuning mainly arises from the unresolved hyperfine structure. By employing AIC to the experimental spectra, it was found that our observed coherent spectral response appeared in the crossover regime between EIT and ATS. The quantum interference effects studied in the ladder system of ultracold polar molecules, as presented in this work, are of crucial importance for microwave-assisted coherent control with multilevel configuration.

Conflicts of interest

There are no conflicts to declare.

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References

- 1 S. E. Harris, J. E. Field and A. Imamoğlu, *Phys. Rev. Lett.*, 1990, **64**, 1107–1110.
- 2 K.-J. Boller, A. Imamoğlu and S. E. Harris, *Phys. Rev. Lett.*, 1991, **66**, 2593–2596.
- 3 M. Fleischhauer, A. Imamoglu and J. P. Marangos, *Rev. Mod. Phys.*, 2005, 77, 633–673.
- 4 S. H. Autler and C. H. Townes, Phys. Rev., 1955, 100, 703-722.
- 5 O. Kocharovskaya, Phys. Rep., 1992, 219, 175-190.
- 6 M. Sähler, S. Knappe, C. Affolderbach, W. Kemp and R. Wynands, *EPL*, 2001, **54**, 323–328.
- 7 O. Kocharovskaya, Y. Rostovtsev and M. O. Scully, *Phys. Rev. Lett.*, 2001, **86**, 628–631.
- 8 M. D. Lukin, Rev. Mod. Phys., 2003, 75, 457-472.
- 9 J. A. Souza, E. Figueroa, H. Chibani, C. J. Villas-Boas and G. Rempe, *Phys. Rev. Lett.*, 2013, **111**, 113602.
- 10 M. A. Quesada, A. M. F. Lau, D. H. Parker and D. W. Chandler, *Phys. Rev. A: At., Mol., Opt. Phys.*, 1987, **36**, 4107–4110.
- 11 R. Garcia-Fernandez, A. Ekers, J. Klavins, L. P. Yatsenko, N. N. Bezuglov, B. W. Shore and K. Bergmann, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2005, **71**, 023401.
- E. Saglamyurek, T. Hrushevskyi, A. Rastogi, K. Heshami and L. J. LeBlanc, *Nat. Photonics*, 2018, **12**, 774–782.
- 13 B. Yan, S. A. Moses, B. Gadway, J. P. Covey, K. R. A. Hazzard, A. M. Rey, D. S. Jin and J. Ye, *Nature*, 2013, **501**, 521–525.
- 14 A. Micheli, G. K. Brennen and P. Zoller, *Nat. Phys.*, 2006, 2, 341–347.
- V. Andreev, D. G. Ang, D. DeMille, J. M. Doyle, G. Gabrielse, J. Haefner, N. R. Hutzler, Z. Lasner, C. Meisenhelder, B. R. O'Leary, C. D. Panda, A. D. West, E. P. West, X. Wu and A. C. M. E. Collaboration, *Nature*, 2018, 562, 355–360.
- 16 D. DeMille, Phys. Rev. Lett., 2002, 88, 067901.
- 17 J. W. Park, Z. Z. Yan, H. Loh, S. A. Will and M. W. Zwierlein, *Science*, 2017, 357, 372–375.
- 18 J. F. Barry, D. J. McCarron, E. B. Norrgard, M. H. Steinecker and D. DeMille, *Nature*, 2014, **512**, 286–289.
- 19 L. Anderegg, B. L. Augenbraun, E. Chae, B. Hemmerling, N. R. Hutzler, A. Ravi, A. Collopy, J. Ye, W. Ketterle and J. M. Doyle, *Phys. Rev. Lett.*, 2017, **119**, 103201.

- 20 S. Truppe, H. J. Williams, M. Hambach, L. Caldwell, N. J. Fitch, E. A. Hinds, B. E. Sauer and M. R. Tarbutt, *Nat. Phys.*, 2017, 13, 1173–1176.
- 21 K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Pe'er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin and J. Ye, *Science*, 2008, 322, 231–235.
- 22 P. K. Molony, P. D. Gregory, Z. Ji, B. Lu, M. P. Köppinger, C. R. Le Sueur, C. L. Blackley, J. M. Hutson and S. L. Cornish, *Phys. Rev. Lett.*, 2014, **113**, 255301.
- 23 T. Takekoshi, L. Reichsöllner, A. Schindewolf, J. M. Hutson,
 C. R. Le Sueur, O. Dulieu, F. Ferlaino, R. Grimm and
 H.-C. Nägerl, *Phys. Rev. Lett.*, 2014, 113, 205301.
- 24 M. Guo, B. Zhu, B. Lu, X. Ye, F. Wang, R. Vexiau, N. Bouloufa-Maafa, G. Quéméner, O. Dulieu and D. Wang, *Phys. Rev. Lett.*, 2016, **116**, 205303.
- 25 K. Bergmann, H. Theuer and B. W. Shore, *Rev. Mod. Phys.*, 1998, **70**, 1003–1025.
- 26 K. Bergmann, H.-C. Nägerl, C. Panda, G. Gabrielse,
 E. Miloglyadov, M. Quack, G. Seyfang, G. Wichmann,
 S. Ospelkaus, A. Kuhn, S. Longhi, A. Szameit, P. Pirro,
 B. Hillebrands, X.-F. Zhu, J. Zhu, M. Drewsen,
 W. K. Hensinger, S. Weidt, T. Halfmann, H.-L. Wang,
 G. S. Paraoanu, N. V. Vitanov, J. Mompart, T. Busch,
 T. J. Barnum, D. D. Grimes, R. W. Field, M. G. Raizen,
 E. Narevicius, M. Auzinsh, D. Budker, A. Pálffy and
 C. H. Keitel, J. Phys. B: At., Mol. Opt. Phys., 2019, 52, 202001.
- 27 J. Deiglmayr, A. Grochola, M. Repp, K. Mörtlbauer, C. Glück, J. Lange, O. Dulieu, R. Wester and M. Weidemüller, *Phys. Rev. Lett.*, 2008, **101**, 133004.
- 28 P. Zabawa, A. Wakim, M. Haruza and N. P. Bigelow, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2011, **84**, 061401.
- 29 J. Banerjee, D. Rahmlow, R. Carollo, M. Bellos, E. E. Eyler, P. L. Gould and W. C. Stwalley, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2012, 86, 053428.
- 30 T. Shimasaki, M. Bellos, C. D. Bruzewicz, Z. Lasner and D. DeMille, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2015, 91, 021401.
- 31 I. C. Stevenson, D. B. Blasing, Y. P. Chen and D. S. Elliott, *Phys. Rev. A*, 2016, **94**, 062510.
- 32 S. Stellmer, B. Pasquiou, R. Grimm and F. Schreck, *Phys. Rev. Lett.*, 2012, **109**, 115302.
- 33 S. Ospelkaus, K.-K. Ni, G. Quéméner, B. Neyenhuis, D. Wang, M. H. G. de Miranda, J. L. Bohn, J. Ye and D. S. Jin, *Phys. Rev. Lett.*, 2010, **104**, 030402.
- 34 S. A. Will, J. W. Park, Z. Z. Yan, H. Loh and M. W. Zwierlein, *Phys. Rev. Lett.*, 2016, **116**, 225306.
- 35 P. D. Gregory, J. Aldegunde, J. M. Hutson and S. L. Cornish, *Phys. Rev. A*, 2016, **94**, 041403.
- 36 M. Guo, X. Ye, J. He, G. Quéméner and D. Wang, *Phys. Rev.* A, 2018, 97, 020501.
- 37 H. J. Williams, L. Caldwell, N. J. Fitch, S. Truppe, J. Rodewald, E. A. Hinds, B. E. Sauer and M. R. Tarbutt, *Phys. Rev. Lett.*, 2018, **120**, 163201.
- 38 Z. Z. Yan, J. W. Park, Y. Ni, H. Loh, S. Will, T. Karman and M. Zwierlein, *Phys. Rev. Lett.*, 2020, **125**, 063401.
- 39 A. V. Gorshkov, S. R. Manmana, G. Chen, E. Demler, M. D. Lukin and A. M. Rey, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2011, 84, 033619.

- 40 S. R. Manmana, E. M. Stoudenmire, K. R. A. Hazzard, A. M. Rey and A. V. Gorshkov, *Phys. Rev. B: Condens. Matter Mater. Phys.*, 2013, 87, 081106.
- 41 B. Sundar, B. Gadway and K. R. A. Hazzard, *Sci. Rep.*, 2018, 8, 3422.
- 42 R. Sawant, J. A. Blackmore, P. D. Gregory, J. Mur-Petit, D. Jaksch, J. Aldegunde, J. M. Hutson, M. R. Tarbutt and S. L. Cornish, *New J. Phys.*, 2020, 22, 013027.
- 43 J. A. Blackmore, P. D. Gregory, S. L. Bromley and S. L. Cornish, *Phys. Chem. Chem. Phys.*, 2020, 27529–27538.
- 44 M. Aymar and O. Dulieu, J. Chem. Phys., 2005, 122, 204302.
- 45 I. M. Georgescu, S. Ashhab and F. Nori, *Rev. Mod. Phys.*, 2014, **86**, 153–185.
- 46 D. J. McCarron, H. W. Cho, D. L. Jenkin, M. P. Köppinger and S. L. Cornish, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2011, 84, 011603.
- 47 E. R. Hudson, N. B. Gilfoy, S. Kotochigova, J. M. Sage and D. DeMille, *Phys. Rev. Lett.*, 2008, **100**, 203201.
- 48 Z. Li, Z. Ji, T. Gong, J. Cao, Y. Zhao, L. Xiao and S. Jia, *Opt. Express*, 2018, 26, 2341–2348.
- 49 Z. Ji, T. Gong, Y. He, J. M. Hutson, Y. Zhao, L. Xiao and S. Jia, *Phys. Chem. Chem. Phys.*, 2020, **22**, 13002–13007.
- 50 P. M. Anisimov, J. P. Dowling and B. C. Sanders, *Phys. Rev. Lett.*, 2011, **107**, 163604.
- 51 D. Wang, J. T. Kim, C. Ashbaugh, E. E. Eyler, P. L. Gould and W. C. Stwalley, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2007, 75, 032511.
- 52 J. Yuan, Y. Zhao, Z. Ji, Z. Li, J. T. Kim, L. Xiao and S. Jia, J. Chem. Phys., 2015, 143, 224312.
- 53 D. M. Brink and G. R. Satchler, *Angular momentum*, Clarendon Press, Oxford, 3rd edn, 1994.
- 54 J. T. Hougen, The Calculation of Rotational Energy Levels and Rotational Line Intensities in Diatomic Molecules, Natl. Bur. Std. (US) Monogr. 115, 1970.
- 55 T. Y. Abi-Salloum, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2010, **81**, 053836.
- 56 J. Gea-Banacloche, Y. Li, S. Jin and M. Xiao, *Phys. Rev. A: At., Mol., Opt. Phys.*, 1995, **51**, 576–584.
- 57 F. Zhou, Y. Niu and S. Gong, J. Chem. Phys., 2009, 131, 034105.
- 58 Atomic physics, ed. C. J. Foot, Oxford University Press, 2004.
- 59 J. Aldegunde and J. M. Hutson, Phys. Rev. A, 2017, 96, 042506.
- 60 J. M. Hutson, (private communication).
- 61 L. Giner, L. Veissier, B. Sparkes, A. S. Sheremet, A. Nicolas,
 O. S. Mishina, M. Scherman, S. Burks, I. Shomroni,
 D. V. Kupriyanov, P. K. Lam, E. Giacobino and J. Laurat, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2013, 87, 013823.
- 62 C. Zhu, C. Tan and G. Huang, *Phys. Rev. A: At., Mol., Opt. Phys.*, 2013, **87**, 043813.
- 63 J. Liu, H. Yang, C. Wang, K. Xu and J. Xiao, *Sci. Rep.*, 2016, 6, 19040.
- 64 B. Peng, S. K. Özdemir, W. Chen, F. Nori and L. Yang, *Nat. Commun.*, 2014, 5, 5082.
- 65 L. He, T. Wang, Y. Gao, C. Cao and C. Wang, *Opt. Express*, 2015, **23**, 23817–23826.
- 66 Q. Liu, T. Li, X. Luo, H. Zhao, W. Xiong, Y. Zhang, Z. Chen, J. S. Liu, W. Chen, F. Nori, J. S. Tsai and J. Q. You, *Phys. Rev. A*, 2016, **93**, 053838.