Measurement of the permanent electric dipole moment of ultracold ground state ⁸⁵Rb¹³³Cs molecules by microwave coherent spectroscopy

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Abstract: We demonstrate measurement of the permanent electric dipole moment (EDM) of ⁸⁵Rb¹³³Cs molecules in the absolute vibrational ground state by microwave (MW) coherent spectroscopy. The rotational states of the considered molecules, which are formed from short-range photoassociation of mixed cold atoms, are nondegenerated under external electric field. To measure the EDM based on electric-field-induced shifts of the sublevels of $X^1\Sigma^+(v=0, J=1)$ rotational state, we utilized a MW coherent spectroscopy and can also eliminate the influence from Stark shift of the excited state existing in both spectroscopies above. In order to acquire accurate electric intensity, electromagnetic induced transparency spectroscopy of ⁸⁵Rb Rydberg atoms is used to implement the calibration. The permanent EDM of ⁸⁵Rb¹³³Cs molecules is finally determined to be 1.266(15) D, which agrees with the theoretical calculations and is comparable with the value of its isotopic molecule.

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1. Introduction

Ultracold polar molecules have opened exciting new research areas based on their long-range and anisotropic dipole-dipole interactions [1,2], including quantum information [3], cold chemistry [4,5] and precision measurement [6,7]. In these applications, the intrinsic permanent electric dipole moment (EDM) plays crucial roles where the dipole-dipole interaction needs to be tuned by external field [8]. Thus the accurate knowledge on EDM of polar molecules is especially important.

Heteronuclear alkali diatomic molecules in the absolute ground state, *i.e.* $X^1\Sigma^+(v = 0)$, are expected to possess EDM range from half to a few Debye in theory [9,10]. Ultracold ground state ⁴⁰K⁸⁷Rb [11], ²³Na⁴⁰K [12], ⁶Li²³Na [13], ⁸⁷Rb¹³³Cs [14,15], ²³Na⁸⁷Rb [16] and ²³Na³⁹K [17] molecules have been formed by magnetoassociation (MA) and subsequently stimulated Raman adiabatic transfer (STIRAP), ⁸⁵Rb¹³³Cs [18,19], ⁷Li¹³³Cs [20], NaCs [21] have been prepared from short-range photoassociation (PA) followed by spontaneously decay. Among these molecules, the EDMs of ⁴⁰K⁸⁷Rb [11], ⁸⁷Rb¹³³Cs [14,15], ²³Na⁸⁷Rb [16] and ⁷Li¹³³Cs [22] have been obtained experimentally by applying an electric field where the induced energy shift or splitting of molecules in ground state are measured.

To our knowledge, there is no report on the EDM measurement of ⁸⁵Rb¹³³Cs molecules, which are attractable for its special characteristics: sizable permanent EDM [9] renders quantum simulation [23]; avoidable immiscibility of its components, which is different from its isotopic components [24], provides possibility to realize molecular Bose-Einstein condensation; inelastic collision with co-trapped Cs is conducive to realize the purification in lowest vibronic state [25]. Experiments showed that short-range PA combined with photoionization (PI) can be exploited

to accomplish the production and detection of ⁸⁵Rb¹³³Cs molecules in ground state [19,26,27]. However, the rotational level can't be distinguished generally because of the broad linewidth of PI laser (around several GHz), let alone its sublevels induced by the electric field. In 2010, Deiglmayr *et al.* utilized a depletion spectroscopy in such kind of experimental setup to realize the resolution of rotational level and achieve the EDM measurement of ⁷Li¹³³Cs molecule [22]. However the measurement uncertainty is large due to the following reasons. The resolution of depletion spectroscopy is on the order of MHz, limited by short lifetime of the coupled excited state (normally tens of nanoseconds). Furthermore, the upper level coupled by depletion laser also has Stark shift under electric field (albeit small), but is ignored by the authors.

By comparing with depletion spectroscopy, microwave (MW) can be used to couple two adjacent rotational quantum states of polar molecules with a higher resolution [28]. In our previous publication [29], we demonstrated that the precision obtained by one-photon MW spectroscopy is almost three orders of magnitude better than the depletion spectroscopy. Recently we further shows that two-photon MW coherent spectroscopy has three times higher resolution than one-photon MW spectroscopy [30]. Thus it is expected to be more accurate to measure EDM if we use MW coherent spectroscopy instead of depletion spectroscopy. Another advantage is that MW coherent spectroscopy can eliminate the influence from Stark shift of the excited state, which exists both in depletion spectroscopy and one-photon MW spectroscopy. In the electric-field-induced lambda-type three-level system, two MW fields couple the same upper level. Thus the frequency difference between these two MW fields exactly represents the Stark splitting between these two lower sublevels, without influence from the upper level.

In this paper, MW coherent spectroscopy is used to measure sublevel splittings of ${}^{85}\text{Rb}{}^{133}\text{Cs}$ molecules in the $X^{1}\Sigma^{+}(v = 0, J = 1)$ rotational state under electric fields, which is calibrated by utilizing the Stark shift of Rydberg ${}^{85}\text{Rb}$ atoms. The EDM of ${}^{85}\text{Rb}{}^{133}\text{Cs}$ molecules in the absolute vibrational ground state is then determined and compared with theoretical calculations and other related experiments.

2. Experimental setup

The production and detection scheme of our experiment is shown in Fig. 1(a-c), similar as Ref. [30], in which two-photon MW spectroscopy is demonstrated. Therefore here we only give main procedures and parameters, especially the information on the response of molecules under an additional electric field. The potential energy curves are based on the data in Refs. [31,32]. The relevant rotational energy levels J and their projections m_J under external field are displayed in Fig. 1(b). The time sequence is shown in Fig. 1(c) and can be divided into three parts, *i.e.* molecules production, coherent interaction and population detection.

In this work, 1×10^{7} ⁸⁵Rb atoms in $5S_{1/2}(F = 2)$ state with a density of 8×10^{10} cm⁻³ and 2×10^{7} ¹³³Cs atoms in $6S_{1/2}(F = 3)$ state with a density of 1.5×10^{11} cm⁻³ are trapped in the dark spontaneous force optical traps. A Ti:sapphire laser, with typical linewidth of 100 kHz and output power up to ~1 W, locked by a subtle transfer cavity technique using an ultrastable He-Ne laser as a reference laser [33], is applied to excite pairs of colliding ⁸⁵Rb and ¹³³Cs atomic samples to photoassociate ultracold ⁸⁵Rb¹³³Cs molecules in $2^{3}\Pi_{0^{-}}(v = 11, J = 0)$ excited state. We choose this state as an intermediate state owing to the completely distribution in J = 1 rotational level for $X^{1}\Sigma^{+}(v = 0)$ state via two-photon cascade spontaneous decay [18], allowing a simple spectroscopy measurement. By applying an additional electric field (named as Stark field) the energy level of $X^{1}\Sigma^{+}(v = 0, J = 1)$ (hereafter called X(0, 1)) will split into sublevels of $|m_{J}| = 0$ and $|m_{J}| = 1$ due to the DC Stark effect (labeled as X(0, 1, 0) and X(0, 1, 1)), where J is the principal quantum number and m_{J} is the projection onto the molecular axis, its absolute value is from 0 to J. MW can be used to couple these sublevels with the selection rules of $\Delta m_{J} = (0, \pm 1)$. We need to notice that it is easier to observe the variation of population if the transition is from a low rotational state to a high rotational state, but harder from a high rotational state to a low



Fig. 1. (a) Optical scheme of production and detection of ${}^{85}\text{Rb}{}^{133}\text{Cs}$ molecules. (b) Rotational energy levels of $X^{1}\Sigma^{+}(v = 0)$ vibrational state in the absence and presence of external electric field. (c) Time sequence.

rotational state. Thus we use transition between X(0, 1) and X(0, 2), rather than X(0, 1) and X(0, 0), to measure the Stark splitting. As mentioned in the introduction, two-photon MW coherent spectroscopy can be used to measure the difference between two lower states via two-photon resonance in a lambda-type three level configuration, regardless of the upper sublevels shift. The relevant energy levels are demonstrated in Fig. 1(b). After turning on the MW field and Stark field for 1ms during the coherent interaction. A continuous wavelength diode laser with linewidth ~MHz, also locked by transfer cavity technique, is used for 2 ms to deplete the population of the rotational state we choose. Here the frequency of depletion laser is chosen to be resonant with the transition from $X^1\Sigma^+(v = 0, J = 2)$ to $B^1\Pi_1(v = 3, J = 1)$. Subsequently, a pulsed dye laser is applied to ionize the molecules in ground state by resonance-enhanced two-photon ionization [34,35]. Finally, the ionzed ⁸⁵Rb¹³³Cs molecules are accelerated to microchannel plate under the action of accelerate electric field where the spacing of two electrode meshes is 4.8 cm. The switches of Stark electric field and accelerate electric field are realized in order by connecting them to two pulsed generators (PVX-4140), followed by a relay.

3. Spectroscopy measurement

Figure 2(a) shows one-photon MW spectroscopy under 2300V electric field by scanning MW frequency. Two peaks correspond to the transition from X(0, 1, 0) to $X(0, 2, |m_J|)$ and X(0, 1, 1) to $X(0, 2, |m_J|)$, respectively. Each experiment point is averaged with 64 times measurements. These two central frequencies are fitted by Lorentz formula to be 1978.48(2) MHz and 1994.93(1) MHz, respectively. We label $|m_J|$ here rather than the specific value because it's not easy to distinguish X(0, 2, 0), X(0, 2, 1) and X(0, 2, 2) due to small splitting of X(0, 2) under the finite electric field intensity we used.

MW coherent spectroscopy can be used to deal with this tricky thing by coupling the same upper level in a lambda-type three-level configuration. The sublevels X(0, 1, 0), X(0, 1, 1) and the combined $X(0, 2, |m_J|)$ can form such a system where $|m_J|$ contains only 0 and 1. We can use two MWs to realize the coupling of these levels. One is the couple field with a fixed frequency, which is chosen to be the transition from X(0, 1, 1) to $X(0, 2, |m_J|)$. The other is the probe field with the scanned frequency around the transition from X(0, 1, 0) to excited state $X(0, 2, |m_J|)$.



Fig. 2. One-photon MW spectroscopy (a) and two-photon MW coherent spectroscopy (b) of ground state 85 Rb 133 Cs molecules under 2300V external electric field.

Figure 2(b) shows the MW coherent spectroscopy in 2300V electric field. We fit the experiment data by using imaginary part of the equation below [36]

$$\chi = \frac{iK}{\gamma_{12} - i\delta_p + \frac{\Omega_c^2/4}{\gamma_{13} - i\delta_p}}.$$
(1)

Here *K* is set to be a coefficient for simplicity in our numerical simulations, γ_{12} and γ_{13} are the decoherence decay rates from $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|1\rangle$ respectively, δ_p is the probe field detuning with the value of $\delta_p = v_p - v_{pc}$, where v_p is the probe field frequency, v_{pc} is the fitting central frequency in Fig. 2(b) and finally fitted to be 1978.49(1) MHz. In the whole measurement we fixed the Rabi frequencies of the coupling field Ω_c as 2.77 MHz and probe field Ω_p as 9 kHz. The difference between X(0, 1, 0) and X(0, 1, 1) can be extracted to be 16.44(1) MHz by subtracting v_{pc} and the fixed coupling frequency.

4. Calibration of electric field intensity

Rydberg atoms, with large principal quantum number $n \ge 10$, can be used to measure the intensity of external electric fields due to their large electric polarizabilities (proportional to n^7) and electric dipole transition matrix elements (proportional to n^2) [37]. In this paper, we adopt the electromagnetic induced transparency (EIT) spectroscopy of ⁸⁵Rb atoms in $10D_{3/2}$ Rydberg state to realize the measurement of the electric field intensity at the molecules position. The suitable Rydberg state is chosen based on these criteria: (1). the coupling laser frequency can cover the transition from intermediate state and Rydberg state; (2). the Stark splitting of the chosen Rydberg state under the electric field should neither be too small to be distinguished by our presented MW coherent spectroscopy, nor too large to induce avoided crossing [38].

A ladder-type three-level system consist of the ground state $5S_{1/2}$, F = 3 ($|g\rangle$), the intermediate state $5P_{3/2}$, F = 4 ($|e\rangle$) and the Rydberg state $10D_{3/2}$ ($|r\rangle$) of ⁸⁵Rb atom. The probe laser is

resonant with the transition from $|g\rangle$ to $|e\rangle$ state. The coupling laser scans around $|e\rangle \rightarrow |r\rangle$ Rydberg transition by a double passed acousto-optic modulators. Probe and coupling beams are overlapped in a counter-propagate configuration and are focused to ultracold ⁸⁵Rb sample with both $1/e^2$ radius of 40 μ m. The powers are around 0.6 μ W and 0.6 mW, respectively. To get EIT spectroscopy, ultracold ⁸⁵Rb atoms are loaded around 90ms firstly and then we turn off the cooling laser and turn on the probe laser and coupling laser with frequency scanning simultaneously for 100 μ s. The repump laser is always turned on to effectively optically pump the F = 2 to F = 3 state.

Figure 3(a) shows the EIT spectrum of ⁸⁵Rb 10D_{3/2} Rydberg atoms we observed. The linewidth is around 15.77(7) MHz with a Lorentz fitting. We attribute the broadened linewidth to the following points: the natural linewidth, coupling laser linewidth and Doppler width, Zeeman broadening of $5P_{3/2}$ state, power broadening and collisional broadening *et al.* When we applied an external electric field, the Rydberg EIT spectrum of ultracold ⁸⁵Rb 10D_{3/2} state splits into two-peak spectra, which correspond to $10D_{3/2,3/2}$ and $10D_{3/2,1/2}$. Figure 3(b) presents the Stark splitting of $10D_{3/2}$ under 1500V electric field. The asymmetric lineshape displayed in Fig. 3(a) and (b) are caused by the radiation pressure of the coupling laser which is similar to the result in Ref. [39]. We use the multi-peak function to fit the experimental data to obtain the central positions of spectrum in which the peak of $10D_{3/2,3/2}$ uses Gauss fitting, the peak of $10D_{3/2,1/2}$ uses Lorentz fitting. By subtracting two central fitting values, we can acquire the difference between $10D_{3/2,3/2}$ and $10D_{3/2,1/2}$. The relationship between the differences and the electric field intensity are shown in Fig. 3(c). The Stark shift of the Rydberg state can be written as [38]

$$\Delta W = -1/2 \cdot \alpha (bE_s)^2. \tag{2}$$



Fig. 3. Measurements of Rydberg EIT spectra as a function of the coupling laser detuning without (a) and with (b) external Stark field. (c) The stark splitting between $10D_{3/2,3/2}$ and $10D_{3/2,1/2}$ under different electric field versus inferred electric field.

In this equation, α is the polarizability. We can obtain α of ⁸⁵Rb 10D_{3/2} for different $|m_J|$ based on the theory of the Atom calculator [40] where the polarizability of $|m_J| = 1/2$ is 942.9704 Hz·cm²/V² and $|m_J| = 3/2$ is 1470.203 Hz·cm²/V², respectively. It should be mentioned that the real electric field intensity felt by atoms and molecules is not equal but proportional to the inferred electric field we applied to meshes (here we label as E_s). Thus the parameter b is introduced as a ratio between them. The experiment data in Fig. 3(c) is fitted by Eq. (2), where α is fixed as the difference between these two sublevel polarizabilities. The actual electric field intensity applied to the electrode meshes is finally fitted as 77.5(2)% E_s .

5. Permanent EDM measurement of ⁸⁵Rb¹³³Cs molecules in the $X^{1}\Sigma^{+}(v = 0)$ state

As previously mentioned, the difference between fitting central in Fig. 2(b) and the frequency of the couple MW field presents the separation between two Stark levels X(0, 1, 0) and X(0, 1, 1). A series of measurements can be made similar to Fig. 2(a) and (b) under different Stark fields. The separations between X(0, 1, 0) and X(0, 1, 1) versus real electric field are eventually obtained in Fig. 4. Combined with the Stark splitting and the real electric field after calibration, we can obtain the EDM of ultracold ground state ⁸⁵Rb¹³³Cs molecules. The Stark shifts are quadratic at low electric fields, the shifts of a rotational level *J* induced by the electric field at weak-field regime can be written as [41]

$$\Delta F = \frac{\mu^2 E_s^2}{h B_\nu} \frac{J(J+1) - 3m_J^2}{2J(J+1)(2J-1)(2J+3)} \qquad J \neq 0.$$
(3)

Here μ is the EDM, E_s is the intensity of DC electric field, B_{ν} is the rotational constant with a value of 0.0166582(3) cm⁻¹ [29]. By fitting the experimental data with Eq. (3), the EDM of ⁸⁵Rb¹³³Cs molecules in the rovibrational ground state is obtained to be 1.266(15) D.



Fig. 4. Stark shift separation between X(0, 1, 0) and X(0, 1, 1) versus real DC Stark field. The experimental data is fitted by Eq. (3), which is displayed with the blue curve.

We list available other theoretical calculations and experimental measurements of EDM of RbCs molecules in ground state for comparison, displayed in Table 1. All these theories didn't distinguish the isotopic molecules. In Ref. [10], pseudopotential configuration interaction calculation is used to calculate EDM of RbCs molecules. In Ref. [42], Ab initio calculation is

Year	EDM(Debye)	Species	method	Ref
1986	1.26	RbCs	Theory	[10]
2005	1.25	RbCs	Theory	[42]
2005	1.237	RbCs	Theory	[9]
2005	1.280	RbCs	Theory	[9]
2005	1.204	RbCs	Theory	[9]
2014	1.17(6)	⁸⁷ Rb ¹³³ Cs	Experiment	[14]
2014	1.225(11)	⁸⁷ Rb ¹³³ Cs	Experiment	[15]
2020	1.266(15)	⁸⁵ Rb ¹³³ Cs	Experiment	Our work

Table 1. EDM measurements of RbCs molecules in ground state.

introduced. In Ref. [9], three different series of calculations are presented. The authors employed contracted Gaussian basis sets to calculate the EDM of RbCs for 1.237 D at first. And then removed the contracted orbital from the basis and introduced the ℓ -dependent ECPs and got the result about 1.280 D. Finally, a basis set and cutoff parameters for Cs were used with the result of 1.204 D. In experiment, the EDM of ⁸⁷Rb¹³³Cs molecules is measured to be 1.17(6) D [14] and 1.225(11) D [15] respectively where the molecules is formed from MA and followed by STIRAP. Thus the measured value of EDM of ⁸⁵Rb¹³³Cs lies in the range of theoretical calculation and is comparable with the values of its isotopic molecule.

6. Summary

In this paper, we report the measurement of sublevel splitting of ultracold ⁸⁵Rb¹³³Cs molecules induced by electric field via MW coherent spectroscopy. Two MW fields couple the same upper level through two-photon resonance, which can expel the influence of splitting in the X(0, 2) state under external electric field. The real electric field applied to the electrode meshes in the vacuum has been calibrated subtly by utilizing the sensitivity of Rydberg atoms to external electric fields. Finally, the permanent EDM of ⁸⁵Rb¹³³Cs molecules in X(0, 1) is measured to be 1.266(15) D, which agrees with the theoretical calculations and is comparable with values of its isotopic molecule.

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