

Transition Dipole Moment Measurements of Ultracold Photoassociated $^{85}\text{Rb}^{133}\text{Cs}$ Molecules by Depletion Spectroscopy *

Juan-Juan Cao(曹娟娟)^{1,2}, Ting Gong(宫廷)^{1,2}, Zhong-Hao Li(李中豪)^{1,2}, Zhong-Hua Ji(姬中华)^{1,2,**}, Yan-Ting Zhao(赵延霆)^{1,2}, Lian-Tuan Xiao(肖连团)^{1,2}, Suo-Tang Jia(贾锁堂)^{1,2}

¹State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006

²Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan 030006

(Received 6 July 2018)

The transition dipole moments (TDMs) of ultracold $^{85}\text{Rb}^{133}\text{Cs}$ molecules between the lowest vibrational ground level, $X^1\Sigma^+$ ($v = 0, J = 1$), and the two excited rovibrational levels, $2^3\Pi_{0+}$ ($v' = 10, J' = 2$) and $2^1\Pi_1$ ($v' = 22, J' = 2$), are measured using depletion spectroscopy. The ground-state $^{85}\text{Rb}^{133}\text{Cs}$ molecules are formed from cold mixed component atoms via the $2^3\Pi_{0-}$ ($v = 11, J = 0$) short-range level, then detected by time-of-flight mass spectrum. A home-made external-cavity diode laser is used as the depletion laser to couple the ground level and the two excited levels. Based on the depletion spectroscopy, the corresponding TDMs are then derived to be $3.5(2)\times 10^{-3}ea_0$ and $1.6(1)\times 10^{-2}ea_0$, respectively, where ea_0 represents the atomic unit of electric dipole moment. The enhance of TDM with nearly a factor of 5 for the $2^1\Pi_1$ ($v' = 22, J' = 2$) excited level means that it has stronger coupling with the ground level. It is meaningful to find more levels with much more strong coupling strength by the represented depletion spectroscopy to realize direct stimulated Raman adiabatic passage transfer from scattering atomic states to deeply molecular states.

PACS: 33.80.-b, 32.70.Cs, 32.80.-t

DOI: 10.1088/0256-307X/35/10/103301

Ultracold molecules have attracted widespread attention in recent years. Compared with homonuclear molecules, heteronuclear (or polar) molecules have unique properties: tunable, long-range and anisotropic dipole-dipole interaction. These properties mean ultracold polar molecules have many important applications, such as ultracold chemistry, quantum computer, quantum simulation and precision measurement.^[1,2]

Up to date, the methods to prepare ultracold molecules include photoassociation (PA) by continuous^[3] and shaped laser,^[4] magentoassociation (MA)^[5] or spin-orbit coupling^[6] from precooled atoms, and direct laser cooling^[7] from preexisting molecules. Especially in recent years the short-range PA, due to the merit of direct formation, continuous accumulation and potential direct stimulated Raman adiabatic passage (STIRAP) transfer from scattering atomic states to deeply molecular states,^[8,9] is verified to be an interesting alternative pathway to produce cold polar molecules in the lowest vibrational ground state ($v = 0$).^[10–17] Among these molecules, $^{85}\text{Rb}^{133}\text{Cs}$ ^[13–16] (hereafter superscript will be omitted) is particularly interesting due to several distinctive merits, including chemical stability, considerable permanent electric dipole moment, and low cost. In addition, the molecule product rate is also considerable. However, the formed mechanism for RbCs molecules decays through a two-photon cascade decay, which is different from a one-step spontaneous decay

for other heteronuclear molecules. This is an obstacle to transferring mixed cooled atoms directly to the deeply bound molecular state. Thus it is meaningful to search for more suitable excited PA states which have both singlet and triplet characters for efficiently producing and transferring molecules from atoms. Recently, Shimasaki *et al.* studied such mixed $2^1\Pi_1$ - $2^3\Pi_1$ - $3^3\Sigma_1^+$ states.^[18] It is found that the product rate is comparative with the previous value of two-photon cascade decay, and the direct spontaneous decay may play an important role due to the mixed characters. Especially for the $2^1\Pi_1$ state, it has mainly a single character and may have stronger coupling with the ground state. Thus we need to measure the corresponding transition dipole moments (TDMs), which represent the strength of transition between the initial and final states, and compare the values for the $2^3\Pi_{0+}$ state.

Many effective methods have been used to measure the TDM of atoms and molecules before, such as two-photon dark resonance spectroscopy,^[19] electromagnetically induced transparency,^[20] and Autler–Townes splitting.^[21] All of these systems involve three or four energy levels. Also, a number of samples are usually needed to obtain a good signal-to-noise ratio (SNR). However, the number of experimentally formed cold molecules is ordinarily on the order of thousands or even fewer, which will limit the SNR based on these methods. Usually a rapid, sensitive photoionization (PI) spectroscopy is used to detect the formed cold

*Supported by the National Key Research and Development Program of China under Grant No 2017YFA0304203, the National Natural Science Foundation of China under Grant Nos 61675120 and 11434007, the National Natural Science Foundation of China for Excellent Research Team under Grant No 61121064, the Shanxi Scholarship Council of China, the 1331KSC, the PCSIRT under Grant No IRT13076, and the Applied Basic Research Project of Shanxi Province under Grant No 201601D202008.

**Corresponding author. Email: jzh@sxu.edu.cn

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molecules with vibrational resolution. To realize the rotational resolution of ground state molecules, a depletion spectroscopy is firstly introduced by Wang *et al.*^[22] and has been widely used in recent years. As the depletion laser couples molecules between the ground state and excited states, it is also suitable to measure the corresponding TDM between them.

In this work, we measure the transition dipole moments for $^{85}\text{Rb}^{133}\text{Cs}$ molecules from the lowest vibrational state, $X^1\Sigma^+$ ($v = 0$) to different excited states, $2^3\Pi_{0+}$ ($v' = 10$) and $2^1\Pi_1$ ($v' = 22$). The former of the excited states can produce ground-state RbCs molecules via two-photon cascade decay while the latter has strong spin-orbit mixing with $2^3\Pi_1$ and $3^3\Sigma_1^+$ states, and thus may have more strong one-step spontaneous decay due to strong coupling with the ground state. The corresponding TDM is derived by fitting the dependence of depletion ratio on both power and irradiated time of depletion laser to the classical absorption formula for a two-level system. The TDM for $2^1\Pi_1$ ($v' = 22$) with a factor of 5 larger than the value for $2^3\Pi_{0+}$ ($v' = 10$) is observed, verifying the much stronger mixing with the ground state.

Most details of the apparatus have been described before, such as in our recent^[23] and previous^[15,16] works. The starting point for this work is a mixture of laser cooled ^{85}Rb and Cs atoms in space-adjustable dark spontaneous force optical traps (dark-SPOTs).^[24] Under a vacuum background pressure of around 3×10^{-7} Pa and a magnetic gradient of around 15 G/cm, we trap mixed atomic clouds with about 1×10^7 ^{85}Rb atoms in the $5S_{1/2}$ ($F = 2$) state with a density of 8×10^{10} cm^{-3} , and about 2×10^7 ^{133}Cs atoms in the $6S_{1/2}$ ($F = 3$) state with a density of 1×10^{11} cm^{-3} . The translational temperature of the mixed atoms is measured by time-of-flight (TOF) imaging to be around 100 μK .

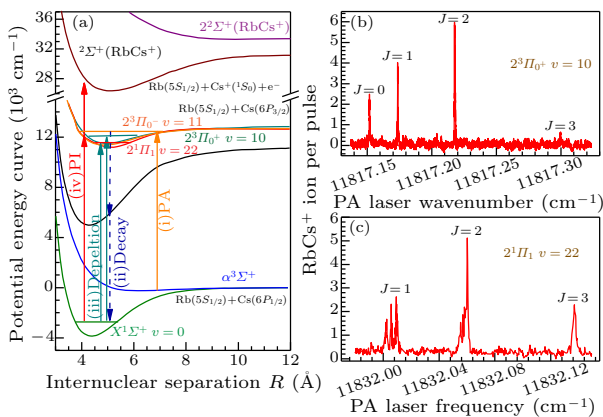


Fig. 1. (Color online) (a) Formation and detection mechanism of ultracold $^{85}\text{Rb}^{133}\text{Cs}$ molecules in the lowest vibrational ground state. (i) PA, (ii) decay, (iii) depletion, and (iv) detection. The potential energy curves are based on the data in Ref. [25]. (b) and (c) are PA spectra for the chosen upper states of depletion transitions, $2^3\Pi_{0+}$ ($v' = 10$) and $2^1\Pi_1$ ($v' = 22$), respectively.

Figure 1(a) shows the preparation and detection mechanisms of formed ultracold $^{85}\text{Rb}^{133}\text{Cs}$ molecules

in the $X^1\Sigma^+$ ($v = 0$, $J = 1$) ground level. (i) First of all, the loading procedure (the duration time of 90 ms) of dark-SPOTs and PA procedure occur simultaneously to produce the molecules in the $2^3\Pi_{0-}$ ($v = 11$, $J = 0$) level. The PA laser (typical linewidth of 100 kHz, output power of 1.5 W, Gaussian radius of 150 μm) is locked by a subtle transfer cavity technique.^[26] (ii) At the same time with (i), the molecules are distributing in different vibrational ground states after the two-photon-cascade spontaneous emission process, including the $X^1\Sigma^+$ ($v = 0$, $J = 1$) level. (iii) Then all the dark-SPOT lasers and PA laser are turned off. Meanwhile, the depletion laser is turned on for several ms to couple the transition from the $X^1\Sigma^+$ ($v = 0$) state to the $2^3\Pi_{0+}$ ($v' = 10$), or $2^1\Pi_1$ ($v' = 22$) state. The depletion laser frequency (linewidth of 2 MHz, actual power of up to 10 mW) can be controlled based on the same transfer cavity technique. To locate the accurate depletion laser frequency, the rotational levels of two upper states are measured with the PA spectrum in advance, as shown in Figs. 1(b) and 1(c). The spectral widths are fitted to be 17(2) MHz and 200(10) MHz by single-peak Lorentz and multi-peaks Lorentz formulas, respectively. The large width for the $2^1\Pi_1$ ($v' = 22$) state arises from the hyperfine structure.^[18] (iv) The PI laser (pulse energy of around 1 mJ, pulse duration of 7 ns, linewidth of 6 GHz, diameter of about 3 mm) with resonant transition between $X^1\Sigma^+$ ($v = 0$) and $2^1\Pi$ ($v = 12$)^[15] is used to photoionize the formed ground-state molecules through one color resonance-enhanced two-photon ionization (RETPI).^[27] The photoionized molecular ions are then accelerated by a pulsed accelerating electric field and detected by a pair of micro channel plates (MCPs). The repetition rate is 10 Hz in our experiment. To obtain depletion spectroscopy, a home-made external-cavity diode laser is scanned around the transitions between the upper and lower molecular levels. Once it is on the resonant, the formed molecules populated in the $X^1\Sigma^+$ ($v = 0$) state will be transferred to other vibrational states, inducing the depletion of molecular ions.

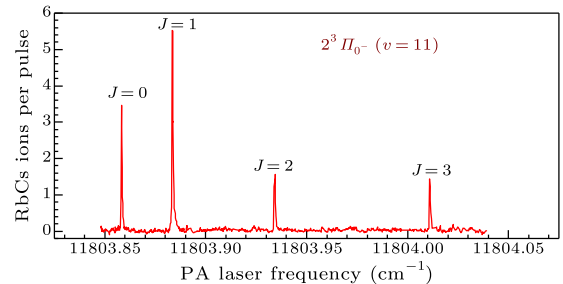


Fig. 2. (Color online) PA spectrum of $^{85}\text{Rb}^{133}\text{Cs}$ molecules in the chosen $2^3\Pi_{0-}$ ($v = 11$) state.

To avoid the same upper state having depletion transition, the PA state here is chosen to be at the $2^3\Pi_{0-}$ ($v = 11$, $J = 0$) level, which also has comparative molecule product rate with $2^3\Pi_{0+}$ ($v = 10$).^[13,16] Figure 2 shows our measurements on the PA spectrum of the $2^3\Pi_{0-}$ ($v = 11$) state, and the photoion-

ized molecular number is truly comparative with the $2^3\Pi_{0+}$ ($v = 10$) observed previously in our experiment. The simple rotational structure is easy for us to assign the rotational number based on our systemic study on the $2^3\Pi_{0-}$ short-range electronic state.^[28] In the following experiments, the PA laser is locked to the $2^3\Pi_{0-}$ ($v = 11, J = 0$) special level because the rotational population is expected to be only $J = 1$ according to Ref. [14], which will benefit the SNR and assignment of depletion spectroscopy. In that study, there is a static electric field left to have Stark mixing inducing that rotational populations are located at $J = 0, 1$, and 2 . However, in our experimental setup, we can eliminate the influence of Stark mixing as the external electric field is pulsed in our experiment. Figures 3(a) and 3(b) show our high resolution depletion spectra of the RbCs molecules in the $X^1\Sigma^+$ ($v = 0$) state. We observe that the depletion is nearly 100% on the transitions from $X^1\Sigma^+$ ($v = 0, J = 1$) to $2^3\Pi_{0+}$ ($v' = 10, J' = 2$) and $2^1\Pi_1$ ($v' = 22, J' = 2$) levels. These results are consistent with the calculations in Ref. [28]. The spectral width in Fig. 3(a) is fitted to be 19(2) MHz while the value in Fig. 3(b) is 180(6) MHz. This is consistent with the PA spectra in Figs. 1(b) and 1(c). To reduce the effect of drifts in the ion signal, we cycle the depletion laser on and off for consecutive detection pulses. The signals with and without the depletion laser are separately average after 192 cycles and a ratio is taken of both the averages to obtain a normalized depletion signal.

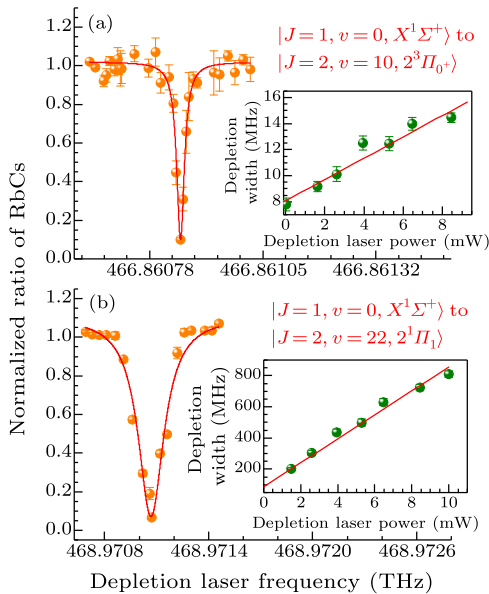


Fig. 3. (Color online) Depletion spectra of $X^1\Sigma^+$ ($v = 0$) state RbCs molecules for the rotational transitions between $J = 1$ to $J' = 2$ of (a) $2^3\Pi_{0+}$ ($v' = 10$) and (b) $2^1\Pi_1$ ($v' = 22$) states. The insets are the dependence of FWHM of depletion spectra on laser power. The solid curves in main graphs are fitted by Lorentz function while the ones in insets are linearly fitted curves.

The upper excited states are chosen to be $2^3\Pi_{0+}$ ($v' = 10, J' = 2$) and $2^1\Pi_1$ ($v' = 22, J' = 2$) levels for Figs. 3(a) and 3(b), respectively, due to the following reasons. The $v' = 10$ vibrational state of the $2^3\Pi_{0+}$

short-range excited state is calculated to have the largest products of free-to-bound and bound-to-bound Frank Condon factors (FCFs)^[13] and also verified in our previous study,^[15] thus one upper excited state of depletion transition is chosen as this vibrational state. As mentioned above, the $2^1\Pi_1$ state is interesting due to the potential large strong coupling with the ground state. In Ref. [18], the vibrational states from $v = 17$ to $v = 34$ are observed. Among these vibrational states, the $v = 22$ state is the best candidate as it is the closest to the strongest photoionized transition, $2^1\Pi_1$ ($v' = 12$) \leftarrow $X^1\Sigma^+$ ($v = 0$),^[13] and also has a considerable molecule product rate. Thus we choose this state as a typical vibrational level of the $2^1\Pi_1$ state. Considering the rotational selection rule $\Delta J = \pm 1$, the upper rotational quantum number is chosen to be $J' = 2$. It is noticed that we can obtain the same measurements when the upper rotational quantum number is chosen to be $J' = 0$ as the rotational quantum state has a slight influence on transition, compared with vibrational quantum states.

This depletion spectroscopy is then used to measure the TDMs between these chosen upper excited states and the ground state, as the rovibrational resonant transitions between the ground state and the excited states can be regarded as an open two-level energy system. The residual molecule number can be expressed as^[29]

$$N = N_0 e^{-t_i \Omega_P^2 \gamma / (\gamma^2 + 4\Delta_P^2)}, \quad (1)$$

where N_0 is the initial number of molecules with far detuning, γ is the linewidth of excited state, t_i is the irradiated time, and Δ_P is the detuning of the laser from resonance. The transition Rabi frequency is $\Omega_P = \langle \mu \rangle E / \hbar$ with $\langle \mu \rangle$ being the TDM, and $E^2 = 4P / \pi \omega^2 c n \epsilon_0$, where E is the electric field vector, P is the depletion laser power, the waist of depletion laser ω is measured to be 0.15 cm, c is the speed of light in a vacuum, n is the refractive index, and ϵ_0 is the permittivity of free space.

When $\Delta_P = 0$, Eq. (1) can be rewritten as

$$N = N_0 e^{-\frac{t_i \Omega_P^2}{\gamma}}. \quad (2)$$

Thus the depletion ratio R can be defined as

$$R = \frac{N_0 - N_0 e^{-\frac{t_i \Omega_P^2}{\gamma}}}{N_0} = 1 - e^{-\frac{4t_i P \langle \mu \rangle^2}{\pi \hbar^2 \omega^2 \gamma c n \epsilon_0}}. \quad (3)$$

According to Eq. (3), $\langle \mu \rangle$ could be deduced when other experimental parameters are determined. Among these parameters, the value of linewidth γ is very important and can be derived from the dependence of full width at half maximum (FWHM) on the depletion laser power, which is shown in the insets of Figs. 3(a) and 3(b). The irradiated times are 2.5 ms and 2 ms, respectively. It is shown that the FWHM increases as the laser power increases. The linewidths are linearly fitted to be 8(1) MHz and 88(5) MHz, respectively.

After we obtain γ , then we can obtain the TDM if we know the values of depletion laser power and irradiated time. To reduce the measured error, we plot the depletion ratio on the power and irradiated time respectively in Figs. 4(a) and 4(b). The irradiated times are 2.5 ms and 2 ms for $2^3\Pi_{0+}$ and $2^1\Pi_1$ states in Fig. 4(a). The powers of depletion laser are 7.3 mW and 1.5 mW for $2^3\Pi_{0+}$ and $2^1\Pi_1$ states, respectively, in Fig. 4(b). We can see that for the two curves, the depletion ratio increases and saturates gradually. We use Eq. (3) to fit Figs. 4(a) and 4(b). According to Eq. (3), we can define the characteristic power $P_c = \pi\hbar^2\omega^2\gamma cn\varepsilon_0/4t\langle\mu\rangle^2$ and characteristic time $t_c = \pi\gamma\hbar^2\omega^2cn\varepsilon_0/4P\langle\mu\rangle^2$. The characteristic powers in Fig. 4(a) are 1.13(8) mW and 0.81(5) mW while the characteristic times in Fig. 4(b) are 0.45(4) ms and 0.98(6) ms for the $2^3\Pi_{0+}$ and $2^1\Pi_1$ states, respectively. From these values, the transition dipole moments $\langle\mu\rangle$ between $X^1\Sigma^+$ ($v = 0$, $J = 1$) and $2^3\Pi_{0+}$ ($v' = 10$, $J' = 2$) are obtained to be $3.6(3)\times 10^{-3}ea_0$ and $3.4(2)\times 10^{-3}ea_0$, where e is the electron charge, a_0 is the Bohr radius, and ea_0 is the atomic unit of electric dipole moment. The averaged value is $3.5(2)\times 10^{-3}ea_0$. The transition dipole moments $\langle\mu\rangle$ between $X^1\Sigma^+$ ($v = 0$, $J = 1$) and $2^1\Pi_1$ ($v' = 22$, $J' = 2$) are obtained to be $1.6(1)\times 10^{-2}ea_0$ and $1.7(1)\times 10^{-2}ea_0$. The averaged value is $1.6(1)\times 10^{-2}ea_0$. The enhancement of TDM with nearly a factor of 5 for the $2^1\Pi_1$ ($v' = 22$, $J' = 2$) excited level means that it has more strong coupling with the ground level.

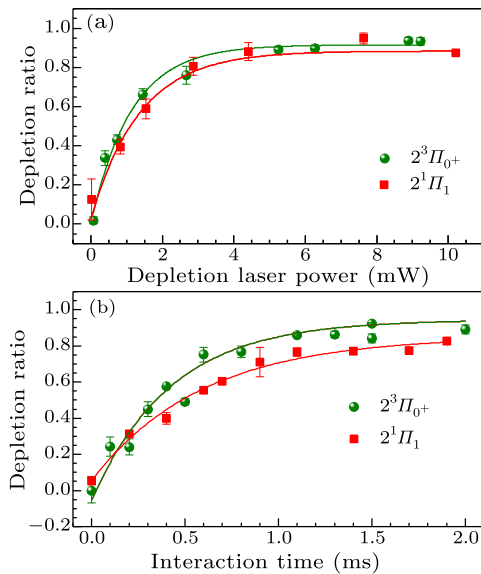


Fig. 4. (Color online) Dependence of the depletion ratio on depletion laser power (a) and irradiated time (b) for rotational transitions from the $X^1\Sigma^+$ ($v = 0$, $J = 1$) level to the $2^3\Pi_{0+}$ ($v' = 10$, $J' = 2$) and $2^1\Pi_1$ ($v' = 22$, $J' = 2$) levels. The curves in (a) and (b) are both fitted by Eq. (3).

In conclusion, we have measured transition dipole moments of ultracold RbCs molecules between the lowest vibrational ground level, $X^1\Sigma^+$ ($v = 0$, $J = 1$), and two excited rovibrational levels, $2^3\Pi_{0+}$ ($v' = 10$,

$J' = 2$) and $2^1\Pi_1$ ($v' = 22$, $J' = 2$) using a depletion spectroscopy. The TDMs are obtained by analyzing the dependence of the depletion ratio on laser power and irradiated time. We need to notice that we measure the TDMs for only one rotational transition between the chosen vibrational states as it is expected and verified that the rotational transition has slight influence on TDMs. However, it is meaningful to find more potential mixed states with stronger coupling strength by the represented depletion spectroscopy to realize direct STIRAP transfer from scattering atomic state to deeply molecular state. In addition, the represented method is also meaningful for other cold molecules.

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