Rotational Population Measurement of Ultracold $^{85}Rb^{133}Cs$ Molecules in the Lowest Vibrational Ground State *

Zhong-Hua Ji(姬中华)[1,2], Zhong-Hao Li(李中豪)[1,2], Ting Gong(宫廷)[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]

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

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

(Received 14 July 2017)

We measure the rotational populations of ultracold $^{85}Rb^{133}Cs$ molecules in the lowest vibrational ground state by a depletion spectroscopy and quantify the molecular production rate based on the measurement of single ion signal area. The $^{85}Rb^{133}Cs$ molecules in the $X^1\Sigma^+(v=0)$ are formed from the short-range $(2)^3\Pi_{0^+}(v=10, J=0)$ molecular state. A home-made external-cavity diode laser is used as the depletion laser to measure the rotational populations of the formed molecules. Based on the determination of single ion signal, the production rates of molecules in the $J=0$ and $J=2$ rotational levels are derived to be 4800 mole/s and 7200 mole/s, respectively. The resolution and quantification of molecules in rotational states are facilitative for the manipulation of rotational quantum state of ultracold molecules.

PACS: 33.80.−b, 37.10.Mn, 32.80.−t, 33.15.Mt DOI: 10.1088/0256-307X/34/10/103301

Ultracold polar molecules have attracted significant interests in the past decades due to their special characteristics, which include permanent electric dipole moment, anisotropic and tunable dipole–dipole interactions.[1] These novel characteristics might facilitate some interesting applications, such as controlled chemistry, precision measurement, quantum computation and quantum simulation.

Researchers have made efforts towards the production of ultracold polar molecules, especially the lowest vibrational ground state (i.e., $X^1\Sigma^+(v=0)$). A pioneering work introduced an incoherent optical pump-dump cycle, where the molecules in the metastable state decay from the photoassociation (PA) level, are excited to another intermediate level and dumped to the $X^1\Sigma^+(v=0)$ state.[2] This scheme is complicated and the total transfer efficiency is low as it needs strong transition probabilities for the related PA laser, pump laser and dump laser. Another elegant and complex control technique, the two-photon stimulated Raman adiabatic passage (STIRAP) process, is used to produce ultracold molecules in special rovibrational ground states starting from Feshbach resonance molecules.[3−7] These transfer processes have a high efficiency and state-selectivity, and the initial cold atomic state is required to be nearly degenerate. However, the molecules can only be formed once per experimental cycle, limiting the accumulation of molecules.

A simple path toward ultracold molecules is short-range PA, where the rovibrational ground state molecules could be produced continuously through direct spontaneous emission after PA. This method has been confirmed in different molecules, including heteronuclear (polar) molecules of $^{39}K^{85}Rb$,[8] $^{23}Na^{133}Cs$,[9] $^{7}Li^{133}Cs$,[10] and $^{7}Li^{85}Rb$.[11] In addition to these heteronuclear alkali molecules, $^{85}Rb^{133}Cs$ is particularly interesting due to several distinctive merits, such as chemical stability, and sizable permanent electric dipole moment. Moreover, compared with the isotope $^{87}Rb^{133}Cs$ molecule, the achievement of Bose–Einstein condensation of $^{85}Rb^{133}Cs$ molecules is feasible as there is no immiscibility between the components.[12]

The efforts to apply short-range PA to $^{85}Rb^{133}Cs$ molecule case were implemented very early.[13−15] However, researchers found that the detected molecules are in the metastable $a^3\Sigma^+$ ground state. Research on intermediate molecular states and absolute ground states is limited. The decay pathways to produce molecules in the $X^1\Sigma^+$ ground state were first calculated through single photon spontaneous decay in Ref. [16] and experimentally verified in Ref. [17] through two-photon cascade decay.[18] According to these calculations and experiments, one appropriate short-range PA state forming absolute ground state molecules is found to be $(5)0^+$ state correlated with the $(2)^3\Pi$ state, which was first investigated in a cold molecular beam[19] and then by PA of cold mixed atoms.[20] The most desired $X^1\Sigma^+(v=0)$ state of $^{85}Rb^{133}Cs$ molecule can be efficiently produced when the intermediated molecules are chosen to be[21] $(2)^3\Pi(v=10)$ and clearly demonstrated by one- and two-color resonance enhanced two-photon ionization (RETPI) in our previous work.[22] However, the $X^1\Sigma^+(v=0)$ state RbCs molecule with vibrational

*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, 11434007 and 61378015, the Program for Changjiang Scholars and Innovative Research Team in University under Grant No IRT13076, and the Applied Basic Research Project of Shanxi Province under Grant No 201601D202008.

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

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resolution cannot distinguish rotational populations as the frequency bandwidth of photoionization (PI) laser is larger than the energy spacing of rotational level.

In this Letter, we measure the rotational populations of the $^{85}Rb^{133}Cs$ molecule in the $X^1\Sigma^+(v=0)$ state using a depletion laser. We also estimate the production rates of resolved $J=0$ and $J=2$ rotational states based on the measurement of single ion signal area. The resolution and quantification of molecules in rotational states will facilitate the manipulation of rotational quantum state of ultracold molecules.

Most details of the apparatus have been described before, such as Ref. [21]. Here we introduce a depletion laser to resolve the rational structures of the formed ground state molecules, use one-color RETPI replacing two-color RETPI as detection mechanism to eliminate the scattering from the 532 nm laser,[22] and quantify the formed molecules based on area measurement of one single ion signal.

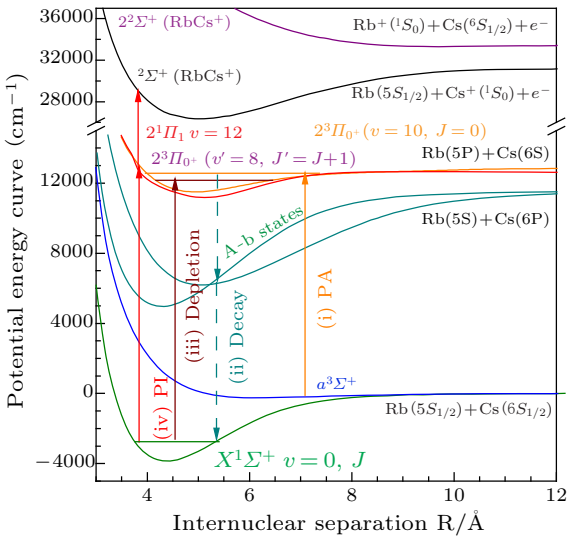

Fig. 1. (Color online) Formation and detection mechanism for ultracold $^{85}Rb^{133}Cs$ molecules in the lowest rovibrational ground state. The $X^1\Sigma^+(v=0)$ state molecules are produced by photoassociating mixed ^{85}Rb-^{133}Cs atoms to the defined intermediate state (i), following the two-photon cascade decay through A-b coupling state (ii). The PI laser is used to ionize the formed molecules with vibrational resolution (iv) while a depletion laser depletes the formed molecules with rovibrational resolution (iii). The potential energy curves are based on the data in Refs. [23,24].

The experiment starts with a mixed ^{85}Rb-^{133}Cs cold sample prepared in our developed space-adjustable dark magnetic-optical trap (MOT).[25] Under a vacuum background pressure of around 3×10^{-7} Pa and a magnetic gradient of around 15 G/cm, we trap a mixed atomic cloud with about 1×10^7 ^{85}Rb atoms in the $5S_{1/2}(F=2)$ state with a density of $8\times10^{10}/cm^3$, and about 2×10^7 ^{133}Cs atoms in the $6S_{1/2}(F=3)$ state with a density of $1\times10^{11}/cm^3$. The translational temperature of the mixed atoms is measured by time-of-flight (TOF) imaging to be around 100 μK.

Figure 1 shows the diagram illustrating the formation and detection mechanisms of ultracold $^{85}Rb^{133}Cs$ molecules in the lowest rovibrational ground state. To form $^{85}Rb^{133}Cs$ molecules in the $X^1\Sigma^+(v=0)$ state, the cold mixed atomic sample is irradiated by a PA laser, which is achieved by employing a tunable Ti:sapphire laser system with a typical linewidth of 100 kHz and an output power of up to 1.5 W. We focus the PA beam on the center of the overlapped dark-MOTs with the Gaussian radius of 150 μm. The PA laser is locked by a subtle transfer cavity technique[26] to a deeply bound level at short range, specifically the $(2)^3\Pi_{0^+}(v=10, J=0)$ rovibrational state which is correlated with the Rb($5P_{1/2}$)+Cs($6S_{1/2}$) atomic asymptote.[20] The uncertainty of locked PA laser frequency can be less than 2 MHz.

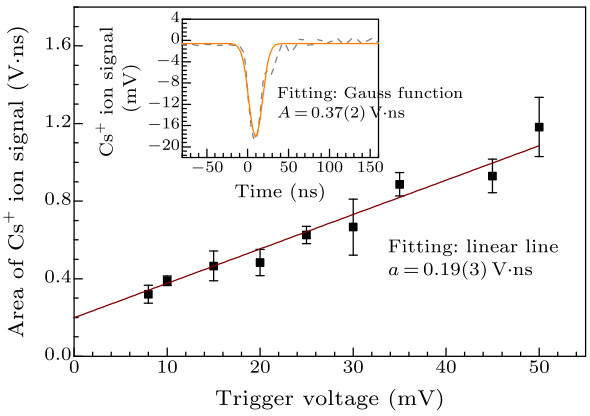

Fig. 2. (Color online) The area measurement of one Cs^+ atomic ion signal. The area is obtained by fitting the atomic ion with a Gaussian function, as shown in the inset.

The formed $X^1\Sigma^+(v=0)$ vibrational ground state molecules via two-photon cascade decay is ionized by a tunable dye laser. The wavelength is fixed to the transition between $X^1\Sigma^+(v=0)$ and $(2)^1\Pi_1(v=12)$ states. The pulse energy is about 1 mJ with a diameter of about 3 mm and a pulse duration of 7 ns. The photoionized molecular ions are then accelerated by a pulsed electric field and detected by a pair of microchannel plates (MCPs). After amplification, the electric signals can be monitored and recorded on a digital oscilloscope, or recorded continuously using an NI PCI-1714 card with 10 averages following a boxcar (Boxcar, SRS-250).

In our previous work, the intensity of atomic and molecular ion is expressed in arbitrary unit (in fact in voltage). To obtain the charge due to one single ionic particle which is necessary to measure the production rate of formed molecules, we record the area (i.e., the area between voltage and time) of Cs^+ atomic ion on an oscilloscope under different trigger levels, as shown in Fig. 2. The error bars correspond to standard deviations in mean value, averaged over 48 repeated experimental measurements. We use a Gaussian function to fit the TOF ion signal, as shown in the inset of Fig. 2. The trigger voltage is 10 mV and

the fitted area (corresponding to the charge) is to be 0.37(2) V·ns. The ringing of the TOF signal appears due to the fact that the component of oscilloscope is not impedance matched with the acquired circuit of MCP. From Fig. 2, we can see that the Cs^+ ion area is linear with the trigger voltage and the fitted area of single ion is 0.19(3) V·ns. It is noted that we need to make sure that only one kind of ions can present when we record the ion signal on the oscilloscope. However, it is found that a few Cs_2^+ molecular ions can be ionized in Cs MOT.[27] Thus we reduce the cooling laser power to a level at which several atomic ions can be ionized. Even the measured sample is of atomic ions, the fitted area is universal for Rb^+, Cs^+ and $RbCs^+$ in our experiment because the ionized charge is the same, resulting in the same achieved kinetic energy, the same induced electrons on MCP, and finally the same transferred voltage on the oscilloscope.

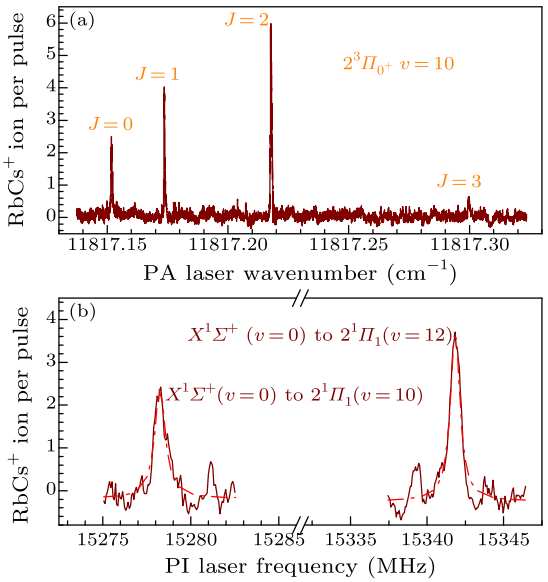

Fig. 3. (Color online) The quantified PA (a) and PI (b) spectra of ultracold $^{85}Rb^{133}Cs$ molecules in the $X^1\Sigma^+(v=0)$ state.

The determination of one single ion allows us to quantify the formed ultracold molecules. Figures 3(a) and 3(b) show the involved PA and PI spectrum of ultracold $^{85}Rb^{133}Cs$ molecules, respectively. The short-range PA state is the mentioned $(2)^3\Pi_{0^+}(v=10)$. We observe in total four rotational levels, in accordance with the observation of Ref. [17], which has an arbitrary unit. The PI spectrum in Fig. 3(b) is obtained by scanning the PI laser frequency while keeping the PA laser locked to the $(2)^3\Pi_{0^+}(v=10, J=0)$ state shown in Fig. 3(a). These transition frequencies between $X^1\Sigma^+(v=0)$ and $(2)^1\Pi_1(v=10)$, $(2)^1\Pi_1(v=12)$ states obtained by one-color RETPI agree with the one obtained by two-color RETPI,[21] but without scattering of the 532 nm laser.

Since the linewidth of the dye laser (~6 GHz) is larger than the spacing of rotational levels, even the largest one between $X(v=0, J=0)$ and $X(v=0, J=1)$, it is impossible to distinguish the rotational level. To resolve the rotational structures, a home-made external-cavity diode laser is used as a depletion laser to excite the formed molecules to another well-known excited molecular state but not active in the RETPI process. The linewidth of depletion laser is the order of MHz and can be scanned or locked by the same transfer cavity technique. The available intensity in our experiment is up to 10 mW and the laser pulse time is chosen to be 2 ms after PA laser.

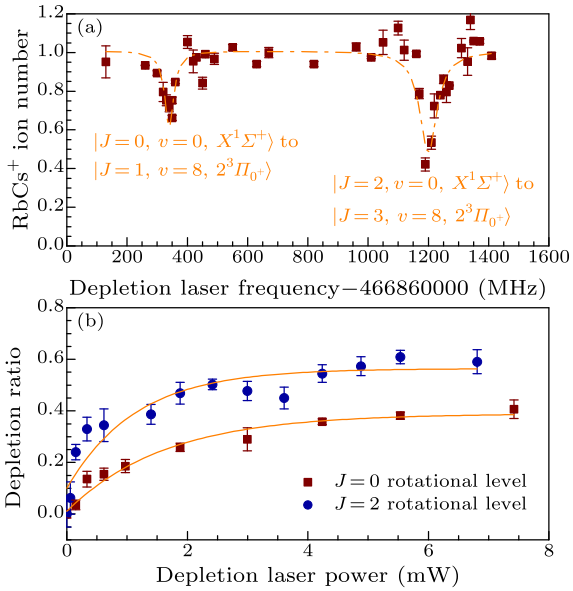

Fig. 4. (Color online) The depletion spectrum (a) and the rotational population measurement (b) of $^{85}Rb^{133}Cs$ in the $X^1\Sigma^+(v=0)$ lowest rovibrational ground state. The dashed curves in (a) are Lorentzian multi-peak fit and the curves in (b) are fitted by Eq. (1). The intensity and pulse time are 2 mW and 2 ms, respectively.

Figure 4(a) shows the obtained depletion spectrum of the formed $^{85}Rb^{133}Cs$ in the $X^1\Sigma^+(v=0)$ lowest vibrational ground state. The depletion laser frequency is scanning around the rotational levels between $X^1\Sigma^+(v=0)$ and $(2)^3\Pi_{0^+}(v'=8)$ states while PA and PI lasers are locked to the $(2)^3\Pi_{0^+}(v=10, J=0)$ state in Fig. 3(a) and the transition between $X^1\Sigma^+(v=0)$ and $(2)^1\Pi_1(v=12)$ states in Fig. 3(b), respectively. Once the depletion laser is on the resonant transition, the distributed population of the resonant rovibrational level in the $X^1\Sigma^+(v=0)$ state can be depleted. The observed ro-vibrational resonant transitions are labeled in Fig. 4(a). Based on this measurement and the previous measured rotational constant of $(2)^3\Pi_{0^+}(v'=8)$ state,[20] the interval of $X^1\Sigma^+(v=0, J=0)$ and $X^1\Sigma^+(v=0, J=2)$ is around 2990 MHz. According to the rigid rotation model, which is especially suitable for low vibrational states, the derived rotational constant of $X^1\Sigma^+(v=0)$ is 499(6) MHz and agrees with the value from the laser-induced fluorescence experiment (497 MHz).[28]

To measure the populations' distribution of rotational states, we need to obtain the saturated depletion ratio. Figure 4(b) shows the dependence of deple-

tion ratio on laser power for the $X^1\Sigma^+(v=0, J=0)$ and $X^1\Sigma^+(v=0, J=2)$ states where the depletion laser is on resonance. By modeling the depletion transition as an open two-level system, the depletion ratio can be expressed by

$$R = R_{\max}(1 - \exp(-\tau_i \Omega^2/\gamma)), \quad (1)$$

where $R_{\max}$ is the maximum depletion ratio, τ_i and Ω are irradiation time and Rabi frequency of the transition. Due to the large noise of ionization detection, we add an offset when we fit our experiment with this equation. The fitting curves in Fig. 4(b) show that the population distributions of $J = 0$ and $J = 2$ states are around 40% and 60%, respectively, with an uncertainty of more than 10%.

Based on the measurements of population distributions and area determination of one single ion signal, the production rate of formed molecules in rotational states could be estimated. The averaged observed $^{85}Rb^{133}Cs^+$ molecular ions are about 3(1) ion/pulse. After turning off the PA laser and dark MOT lasers, the formed molecules begin to expand. The characteristic time τ_c is observed to be around 10 ms, which is larger than the value of Cs_2 in MOT[27] due to low background collision of dark MOT.[29] As the ionization laser arrives at the molecular sample later by 2 ms from the expanding of the molecular sample, the molecular ion at stable status N_{ion} is derived to be 3.7 ion/pulse. The production rate R_{cm} reads[30]

$$R_{\text{cm}} = N_{\text{ion}}/(\tau_c \eta), \quad (2)$$

where η is the gross efficiency. It includes the efficiencies of the ionization process and the detection process, and is estimated to be around 3(1)% as before,[27] which is the same order with other similar apparatus.[30,31] Based on this equation, we obtain that the production rate of $^{85}Rb^{133}Cs$ molecules in the $X^1\Sigma^+(v=0)$ state is $1.2(4) \times 10^4$ mole/s, which is agreeable with the estimated value of our group[21] and Yale group.[17] As the population distributions of $J = 0$ and $J = 2$ states are around 40% and 60%, respectively, their production rates are around 4800±1600 mole/s and 7200±2400 mole/s.

In summary, we have measured the population distributions of rotational levels of $^{85}Rb^{133}Cs$ in the lowest ground state using a depletion spectroscopy, and quantify the production rates for the formed $X^1\Sigma^+(v=0, J=0)$ and $X^1\Sigma^+(v=0, J=2)$ rotational levels based on the determination of one single ion. The distinguishing and quantification of rotational states will facilitate the further research on the manipulation of rotational quantum state, such as microwave transfer and superposition state production of ultracold polar molecules.

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