Photoassociation spectra of ultracold $^{85}Rb_2$ molecule in 0_u^+ long range state near the $5S_{1/2} + 5P_{1/2}$ asymptote*

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We investigate the high resolution photoassociation spectra of $^{85}Rb_2$ molecules in 0_u^+ long range state below the $(5S_{1/2} + 5P_{1/2})$ asymptote. The ^{85}Rb atomic samples are trapped in a dark magneto–optical trap (MOT) and prepared in the dark state. With the help of trap loss technique, we obtain considerable photoassociation spectroscopy with rovibrational resolution, some of which have never been observed before. The observed spectrum is fitted by a rigid rotation model, and the rotational constants of ultracold $^{85}Rb_2$ molecule in long range 0_u^+ are obtained for different vibrational states. By applying the LeRoy–Bernstein method, we assign the vibrational quantum numbers and derive C_3 coefficient, which is used to obtain the potential energy curve.

Keywords: photoassociation spectroscopy, ultracold molecule, C_3 coefficient

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1. Introduction

The theoretical and experimental studies of ultracold molecules have received considerable attentions over the past few decades.[1–4] After successful realization of cooling and trapping ultracold atoms by laser, the researchers begin to focus on ultracold molecules, which have applications in the high resolution molecular spectroscopy,[5] ultracold chemistry,[6] measurement of fundamental physical constants,[7] quantum information and quantum computing.[8]

There are two primary methods used to produce molecules in the sub-milli-kelvin temperature range, which are magnetically tunable Feshbach resonance[9] and light-assisted photoassociation (PA).[10] Although the Feshbach resonance is a highly sufficient method to produce molecules in vibrational levels of the electronic ground state, it has a high demand on the atomic sample. On the contrary, PA technique has following advantages: low requirements of initial temperature of atomic sample, continuous formation of a large number of molecules, simple experimental operation. Thus it is widely used to prepare the ultracold molecules of alkali metal elements and alkaline earth metal elements.[11,12] At the same time, the ultracold excited diatomic molecules produced by the PA technique usually have large internuclear distances, which provide the possibility for studying the properties of excited long range molecules.

PA can be described as this process $A + B + h\nu \rightarrow AB^*$, in which a colliding pair of atoms A and B absorb a photon of frequency ν to produce a bound excited molecule AB^*. Long range molecular states can be efficiently formed because the wave functions of initial collisional states have large amplitudes at long range. These states are expected to exhibit a variety of interesting behaviors related to the recoupling of atomic angular momenta at large atomic separation and to retardation effects.[13–15] PA spectroscopy of cold atoms can also provide information on the long range interactions between atoms, which is necessary to observe Bose–Einstein Condensation (BEC),[16] and other cold atom experiments. Many experimental and theoretical work have studied the long range states of ultracold molecules produced by PA. Ratliff *et al.* studied spectra of ultracold Na_2 molecules in 0_g^-, 1_g, 0_u^+ states and obtained the corresponding rotational constants.[17] Burke *et al.* analyzed spectra of ultracold $^{39}K_2$ molecule in 0_g^- state and measured the scattering length of singlet state and triplet state.[18] Pichler *et al.* studied PA spectra of ultracold Cs_2 molecular in 0_g^-, 1_g, 0_u^+ states and obtained the rotational constants and C_3 coefficient.[19] Ma *et al.* extended the Cs_2 molecular PA spectrum in the 0_u^+ state over 60 cm^{-1} below $6S_{1/2}$+$6P_{3/2}$ dissociation limit.[20] Bergeman *et al.* made a study of the Rb_2 molecules, and gave a more comprehensive 0_u^+ state spectral data.[21] The rotational constants obtained by experimental fitting are meaningful for estimating the unobserved molecular energy levels and obtaining the molecular potential energy curves.

In this paper, we report the photoassociative production of the ultracold $^{85}Rb_2$ molecules correlated to $(5S_{1/2} + 5P_{1/2})$

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dissociation. By trap loss spectroscopy, we observe some rovibrational levels which exist in theory but have not been observed before. According to a rigid rotor model,[22] we analyze the PA spectra and obtain corresponding rotational constants, which are consistent with the theoretical calculations. By applying the LeRoy–Bernstein method,[23] we assign the vibrational quantum numbers and derive C_3 coefficient, which is used to obtain the potential energy curves.

2. Experimental setup

Figure 1 shows the PA process of ultracold $^{85}Rb_2$ molecule. In a PA process, a pair of free cold atoms absorbs one photon with a resonant wavelength to form an excited molecule. This molecule may decay by spontaneous emission to two atoms, which have higher kinetic energy and leave the atomic cloud rapidly, or may form a translational cold molecule in singlet or triplet ground states. In either of these passages, the trapped cold atoms will loss at the resonant frequency of excited molecular state.

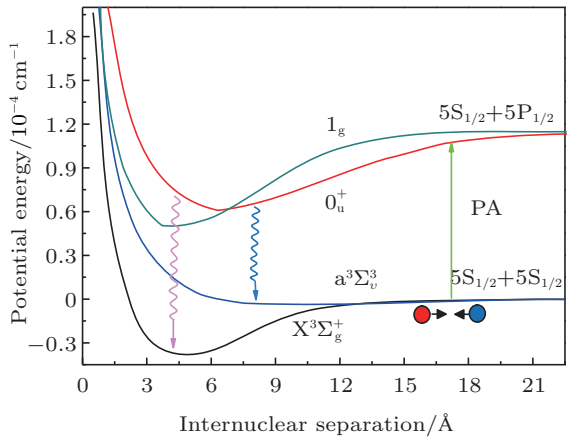


Fig. 1. (color online) Formation and detection schemes of ultracold $^{85}Rb_2$ molecules. Two colliding ultracold $^{85}Rb(5S_{1/2}, F = 2)$ atoms are photoassociated into 0_u^+ state correlated to the $(5S_{1/2} + 5P_{1/2})$ atomic asymptote.

The experimental setup is shown in Fig. 2. We use a dark magneto–optical trap (MOT)[24] with a background pressure of about 1×10^{-7} Pa to trap cold ^{85}Rb atoms. A pair of coils generates a magnetic gradient of about 15 Gs/cm (1 Gs = 10^{-4} T) and three other pairs of coils generate a compensate geomagnetism at the position of cold atom clouds. The trapping and repumping beams are provided by two Littrow external-cavity diode lasers (DL pro, Toptica). The trapping beam is locked to the ^{85}Rb atomic hyperfine transition $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 3)$ by saturated absorption spectroscopy technique. The acousto–optic modulator (AOM) will shift laser frequency to the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ hyperfine transition with the red detuning of 15 MHz. Each trap beam is 20 mm in diameter and 8 mW in power. The repumping frequency is locked to the ^{85}Rb atomic hyperfine resonance transition line $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$, and the power is about 5 mW. The depumping laser frequency is lacked to the transition line $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ by AOM and the power is around 40 μW. The number of ultracold ^{85}Rb atoms in the sample is about 1×10^7, the atomic temperature is about 100 μK. Atomic clouds are observed by two charge-coupled device (CCD) cameras placed along the horizontal and vertical directions.

The PA transition is driven by a Ti:sapphire laser with a typical linewidth of less than 100 kHz and output power of up to ∼ 1.5 W. This beam is focused on the atomic clouds with a Gaussian diameter of 800 μm, which can completely cover the ultracold atom clouds. The frequency of PA is monitored by a commercial wavelength meter (WS/7R) with an absolute accuracy of 60 MHz. In order to ensure the accuracy of frequency measurement, the wavelength meter is calibrated by an He–Ne laser (HRS015B, Thorlabs).

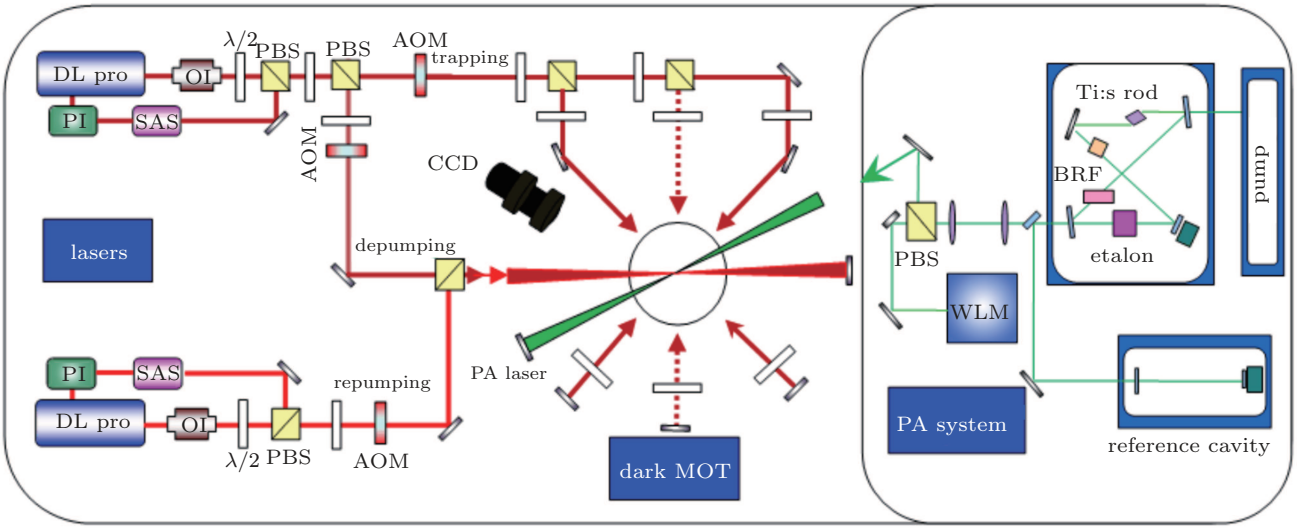


Fig. 2. (color online) Experimental setup. SAS: saturated absorption spectroscopy, AOM: acousto–optic modulator, PBS: polarization beam splitter, and WLM: wavelength meter.

3. Results and discussions

We have observed rovibrational levels of $^{85}Rb_2$ molecules in the 0_u^+ electric state correlated to the $(5S_{1/2} + 5P_{1/2})$ asymptote. Figure 3 shows a typical PA spectrum of $v = 186$ vibrational level, which was not observed before.

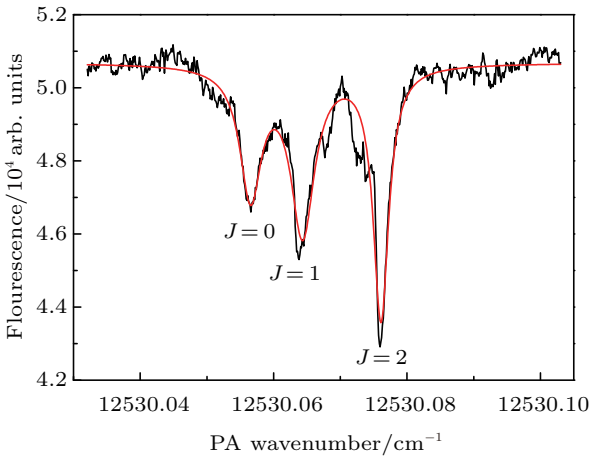


Fig. 3. (color online) A typical PA spectrum of the 0_u^+, $v = 186$ vibrational level.

Bergeman *et al.*[21] measured several resonances of 0_u^+ state of $^{85}Rb_2$ molecules by the trap loss method, but they did not assign vibrational quantum numbers of these states. Based on the previous experimental fitting analysis and experimental observation, the PA frequency in our experiment is scanned down to 12541.102 cm^{-1} from the $(5S_{1/2} + 5P_{1/2})$ asymptote (12579.020 cm^{-1}) to investigate the PA spectroscopy of 0_u^+ long range state. The spectrum is obtained by scanning PA laser frequency at a speed of 30 MHz/s. The PA spectrum is fitted with Lorentz model[25,26] to gain the frequency values of all rotational levels.

According to a nonrigid rotation model,[27] molecular rovibrational energy can be expressed as

$$E_v/hc = B_v J(J+1) - D_v J^2(J+1)^2 \quad (J \geq 0), \qquad (1)$$

where E_v is rovibrational energy eigenvalue, h is Planck's constant, c is the speed of light, v is vibrational quantum number, and J is rotational quantum number, B_v is rotational constant of the v vibrational state, and D_v is the corresponding centrifugal distortion constant and can be ignored for typically two or three orders smaller than B_v in magnitude.

For the same vibrational quantum number, the interval of neighboring rotional levels ΔE can be expressed as

$$\Delta E = [E_v(J) - E_v(J-1)]/hc = 2B_v J \quad (J \geq 1). \qquad (2)$$

With experimental results, the rotational constant can be obtained by Eq. (2). Table 1 gives the transition frequency of different vibrational states at $J = 0$ level. The absolute accuracy of the detuning is 60 MHz. It comes from the accuracy of the wavemeter used in our experiment. However, the relative accuracy of the frequency measurement is much less than this value and is estimated to be 3 MHz. For comparison, we list the values of the corresponding vibrational states measured in Ref. [21]. They observed several resonances by the trap loss method, but there are still many missing states according to theoretical expectations. They attribute the missing observations to the low Frank–Condon factor (FCF)[28] or the low signal to noise ratio of the trap loss method.

Table 1. The frequency values and rotational constants of vibrational state observed by this work and Ref. [21]. Column 1 gives the vibrational number. Column 2 lists the PA laser energy for the $J = 0$ state. Column 3 gives the value of 10^3 B_v . The units of energy and B_v are cm^{-1}.

	This work		Ref. [21]	
v	$E_{L,J=0}$	10^3 B_v	$E_{L,J=0}$	10^3 B_v
v^*	12541.102	3.16	12541.082	3.04
v^*+1	12540.216	1.29		
v^*+2	12538.999	2.56	12538.980	2.40
v^*+4	12536.376	3.84		
v^*+7	12531.763	3.85		
v^*+8	12530.056	2.77		
v^*+9			12528.143	2.44
v^*+10	12526.250	3.16	12526.233	2.76
v^*+11	12524.822	4.54	12524.807	4.47
v^*+12	12523.610	3.30	12523.594	3.20
v^*+13	12521.631	2.04	12521.612	2.60
v^*+14	12519.461	2.17	12519.447	2.69
v^*+15	12517.402	3.93	12517.393	3.45
v^*+16	12516.193	3.79		
v^*+17	12514.394	2.66	12514.375	2.93

The high resolution PA spectra with the rich rotational structures enable us to obtain the molecular constants. Figure 4(a) shows the rotational constants of the vibrational states in Table 1. The experimental measurements of two groups show agreement with each other. The rotational constants can be expressed as $B \approx h/8\mu\pi^2 cR^2$, where μ is reduced mass, R is nuclear interval between two atoms. Rotational constant changes little for that the nuclear interval of vibrational state varies little near the dissociation.

The $^{85}Rb_2$ molecule in excited state consists of S ground state and P excited state of ^{85}Rb atoms, the long range part of molecular state potential energy curves can be approximated as $-C_3/R^3$, where C_3 is excited long term coefficient. In order to obtain the C_3 coefficient, we used the LeRoy–Bernstein (LRB) formula[23]

$$\nu_D - \nu = \frac{\sqrt{2\mu}(-C_3)^{1/3}}{\hbar\sqrt{\pi}} \frac{\Gamma(1/2+1/3)}{\Gamma(1+1/3)} (D - E_v)^{1/6}, \qquad (3)$$

where Γ is the gamma function, $\hbar$ is the reduced Planck constant, D is the dissociation energy, E_v is the energy of the v-th

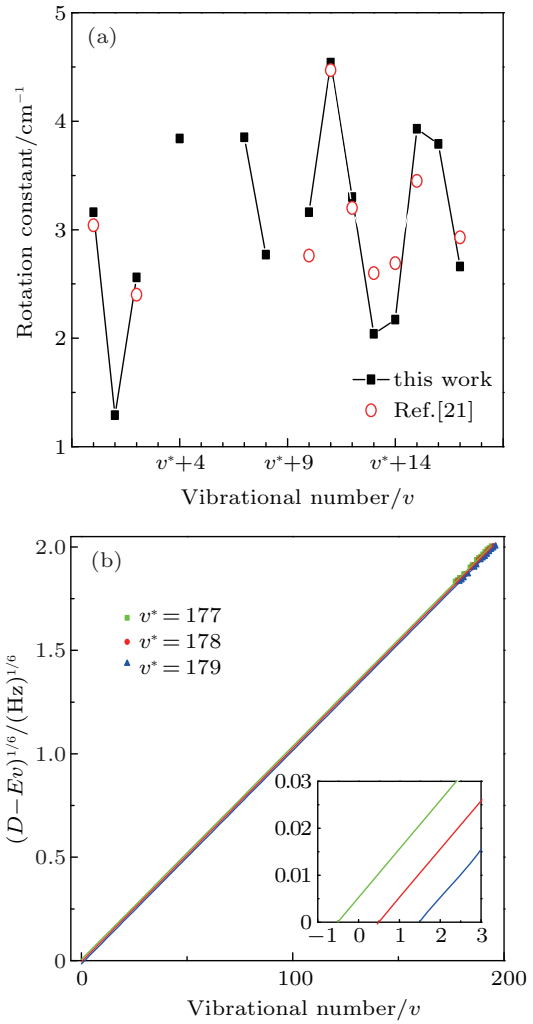


Fig. 4. (color online) (a) The rotational constants of all observed vibrational quantum states in Table 1. (b) The fitting of the experimental results by LRB formula in Eq. (3) with different v^*. The three linear fittings can extract the same C_3 coefficient, but different v_D. The inset is a zoom near horizontal axis.

vibrational level at $J = 0$ rotational state. Based on Eq. (3) and the data in Table 1, we can deduce the values of C_3. The v^* of the data in Table 1 represents an positive integer and is determined below. We vary the values of v^* and can obtain the corresponding values of v_D, which should be in the range of 0 and 1. Figure 4(b) shows the perfect linear fitting of the experimental data with different v^*. The inset is a zoom near horizontal axis. The values of v_D are −0.504, 0.496, and 1.496 for $v^* = 177$, 178, and 179. Only the value of $v^* = 178$ satisfies this condition. So we can determine the exact value of v^* to be 178. For the 0_u^+ state, the fitted value of C_3 is 8.883 ±0.083 a.u. The uncertainty is mainly due to frequency determination based on wavelength meter and experimental data fitting. The theoretical value of C_3 is calculated to be 9.202 a.u., 9.400 a.u., and 10.06 a.u. by Refs. [29], [30], and [31], respectively. The C_3 is often used to obtain the long range part of molecular potential energy curve. In the LeRoy–Bernstein method, the observed successive vibrational levels are assumed to be due to a long range molecular potential energy curve behaving asymptotically as $-C_3/R^3$. In terms of $V(R) = -C_3/R^3$, the empirical potential energy curve for the 0_u^+ long range state below ^{85}Rb ($5S_{1/2} + 5P_{1/2}$) asymptote is shown in Fig. 5. The different curves are derived from the experimental values of C_3 (blue line) and theoretical values of Ref. [29] (red line), Ref. [30] (green line), and Ref. [31] (black line), respectively. It can be seen that the potential energy curve based on the experimental value is close to the red line, meaning that it is more accurate. Our measurements also give a good revise of the corresponding theory and are helpful for studying the potential curve near asymptote for other excited states or other homonuclear molecules.

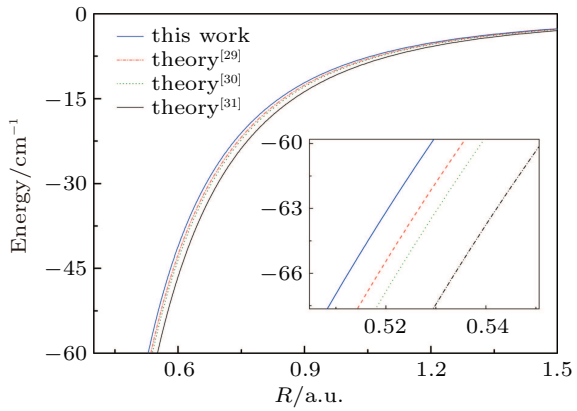


Fig. 5. (color online) The long range parts of potential energy curves for 0_u^+ long range state based on the experimental values of C_3 (blue line) and theoretical value of Ref. [29] (red line), Ref. [30] (green line), and Ref. [31] (black line). The value of zero in vertical coordinate represents the ^{85}Rb ($5S_{1/2} + 5P_{1/2}$) asymptote (12579.020 cm^{-1}).

4. Conclusion

In conclusion, we report high resolution photoassociation spectroscopy of $^{85}Rb_2$ molecules in 0_u^+ long range state below ^{85}Rb ($5S_{1/2} + 5P_{1/2}$) asymptote by high resolution trap loss spectroscopy. With the help of trap loss spectroscopy technique, we obtain considerable high resolution photoassociation spectrum with rovibrational states, some of which have never been observed before. By applying the LeRoy–Bernstein method we assign the vibrational quantum numbers of the 0_u^+ long range state photoassociation spectroscopy when PA frequency is scanned up to 65 cm^{-1} below the ^{85}Rb ($5S_{1/2} + 5P_{1/2}$) asymptote from resonance. The C_3 coefficient is also deduced from the analysis. The curves of our results and theory calculations are almost identical. The derived C_3 coefficient is used to obtain the useful long range molecular potential energy curve. Our experimental results and analytical method are also meaningful for other homonuclear molecules.

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