The laser-intensity dependence of the photoassociation spectrum of the ultracold $Cs_2(6S_{1/2}+6P_{1/2})\ 0_u^+$ long-range molecular state*

Jin Li(金 丽), Feng Guo-Sheng(冯国胜), Wu Ji-Zhou(武寄洲), Ma Jie(马 杰), Wang Li-Rong(汪丽蓉)†, Xiao Lian-Tuan(肖连团), and Jia Suo-Tang(贾锁堂)

State Key Laboratory of Quantum Optics and Quantum Optics Devices, Laser Spectroscopy Laboratory, Shanxi University, Taiyuan 030006, *China*

(Received 22 November 2012; revised manuscript received 10 January 2013)

The high-resolution photoassociation spectrum of the ultracold cesium molecular 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ limit is presented in this paper. The saturation of the photoassociation scattering probability is observed from the dependence of the trap-loss probability on the photoassociation laser intensity. The corresponding resonant line width is also demonstrated to increase linearly with increasing photoassociation laser intensity. Our experimental data have good consistency with the theoretical saturation model of Bohn and Julienne [Bohn J L and Julienne P S 1999 *Phys. Rev. A* **60** 1].

Keywords: photoassociation spectrum, intensity dependence, high resolution

PACS: 87.64.kv, 42.62.Fi, 33.20.Vq, 33.70.Jg **DOI:** 10.1088/1674-1056/22/8/088701

1. Introduction

Photoassociation (PA) of cold atoms has been used as a versatile tool for the formation of ultracold molecules in the past few decades.[1–4] In contrast to molecule formation using Feshbach resonances, PA allows the population of specific rovibrational levels, even those deeply bound in the molecular potentials. In the PA process, starting with pre-cold atoms in the cesium magnetic optical trap (MOT),[5] two colliding ground-state atoms absorb a photon and form an electronically excited molecule at certain rovibrational levels with rather large internuclear separations. The excited molecule has two endings: one is spontaneous decay to form a pair of free atoms, and the other is the transition to ground state to form an ultracold ground-state molecule that has a small fractional probability. So far, PA spectra have been demonstrated for alkali atoms, alkaline earth atoms, and metastable helium atoms.[2] The PA spectra of ultracold atoms have provided significant information about the ground-state collision properties[6,7] between alkali-metal atoms, accurate measurements of the molecular parameters, the PA rate coefficients, and the radiation lifetimes.[8] PA experiments have also given us knowledge of the molecular potentials of the long-range excited state, from which Franck–Condon factors can be calculated, and have allowed us to determine the scattering lengths. Recently, PA studies have been extended to the research of inelastic molecule–atom scattering at ultracold temperatures in heteronuclear dimers, controlled with ultrashort or shaped laser pulses.[9]

The investigation of the dependence of PA probability on laser intensity is of great interest for understanding ultracold collisions and determining the scattering wave function and scattering length. It is a determinative step in the overall molecular formation rate. In the past decades, several investigations of the PA rate have been performed in various samples. McKenzie *et al.*[10] measured the PA spectra for various intensities and durations of the light pulse in a sodium Bose–Einstein condensate, and obtained the limit of the PA rate coefficient, the line shape, and the shift in resonance. Schloder *et al.*[11] studied the intensity dependence of the hyperfine-resolved singlet spectrum in a ^{6}Li^{7}Li two-species MOT. Pordan *et al.*[12] also found that the PA rate is saturated at the unitarity limit of the inelastic scattering cross section of a ^{7}Li Bose–Einstein condensate. Kraft *et al.*[13] then showed the saturation of cesium molecule photoassociation in an optical dipole trap. Mickelson *et al.*[14] investigated the saturation effects in the PA spectrum of ^{86}Sr in 2006. As mentioned above, these studies have presented the laser intensity dependences of the PA rate coefficient in various alkali molecules. However, a systemic measurement of the intensity dependence of the PA rate of the 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ limit has not been performed yet because the 0_u^+ state does not have a special double well structure and the 0_g^- state to manipulate the molecule better in the potential curves.

In this paper, we present the PA spectrum of the 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ dissociation limit with high-resolution in a cesium MOT by monitoring the fluorescence of trapped

*Project supported by the National Basic Research Program of China (Grant No. 2012CB921603), the 863 Program (Grant No. 2011AA010801), the National Natural Science Foundation of China (Grant Nos. 61008012, 10934004, 60978001, 60978018, and 11174187), the International Science & Technology Cooperation Program of China (Grant No. 2011DFA12490), the National Natural Science Foundation of China for Excellent Research Team (Grant No. 61121064), the Natural Science Foundation of Shanxi Province, China (Grant Nos. 2011011004 and 2011081030), and the New Teacher Fund of the Ministry of Education of China (Grant No. 20101401120004).

†Corresponding author. E-mail: wlr@sxu.edu.cn

© 2013 Chinese Physical Society and IOP Publishing Ltd

atoms as a function of PA laser frequency. The dependences of both the PA probability and the line width of the resonant signals on the laser intensity are also analyzed. Our experimental data can be fit to the theoretical saturation model, which is based on the multichannel characteristics of the PA process.[15] The intensity dependences of the PA probability and the line width for different rotational levels at the $v = 169$ vibrational level of the Cs_2 long-range 0_u^+ state are presented, and the difference in the saturation intensity between different rotational progressions are also analyzed.

2. Experimental setup

The experimental setup has been presented in our previous paper.[16] Our experiment was performed in a vapor-cell MOT (see Fig. 1). The MOT is operated in a mutually orthogonal six-beam configuration with opposite $\sigma^{+/-}$ polarizations of an intensity of nearly 3 mW/cm^2. The magnetic field of the trap was provided by a pair of anti-Helmholtz coils with a gradient of 15 Gs/cm (1 Gs = 10^{-4} T) along the axis of the coils. A sample of ultracold Cs atoms was loaded in the trap with a background pressure of about 10^{-7} Pa. The trapping laser frequency was red detuned from transition $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$ by about two natural line widths, while the repumping laser was $6S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$. Both the trapping beam and the repumping beam were produced from a frequency-stabilized diode laser system (DL100). In our experiment, the temperature of the sample was about 200 μK, obtained by the time-of-flight method. The atomic number was measured to be about 5×10^7 using the absorption method, and the dimension of the cold atom cloud was about 700 μm, as measured by a charge-coupled device (CCD) camera, resulting in available intensities up to 150 W/cm^2.

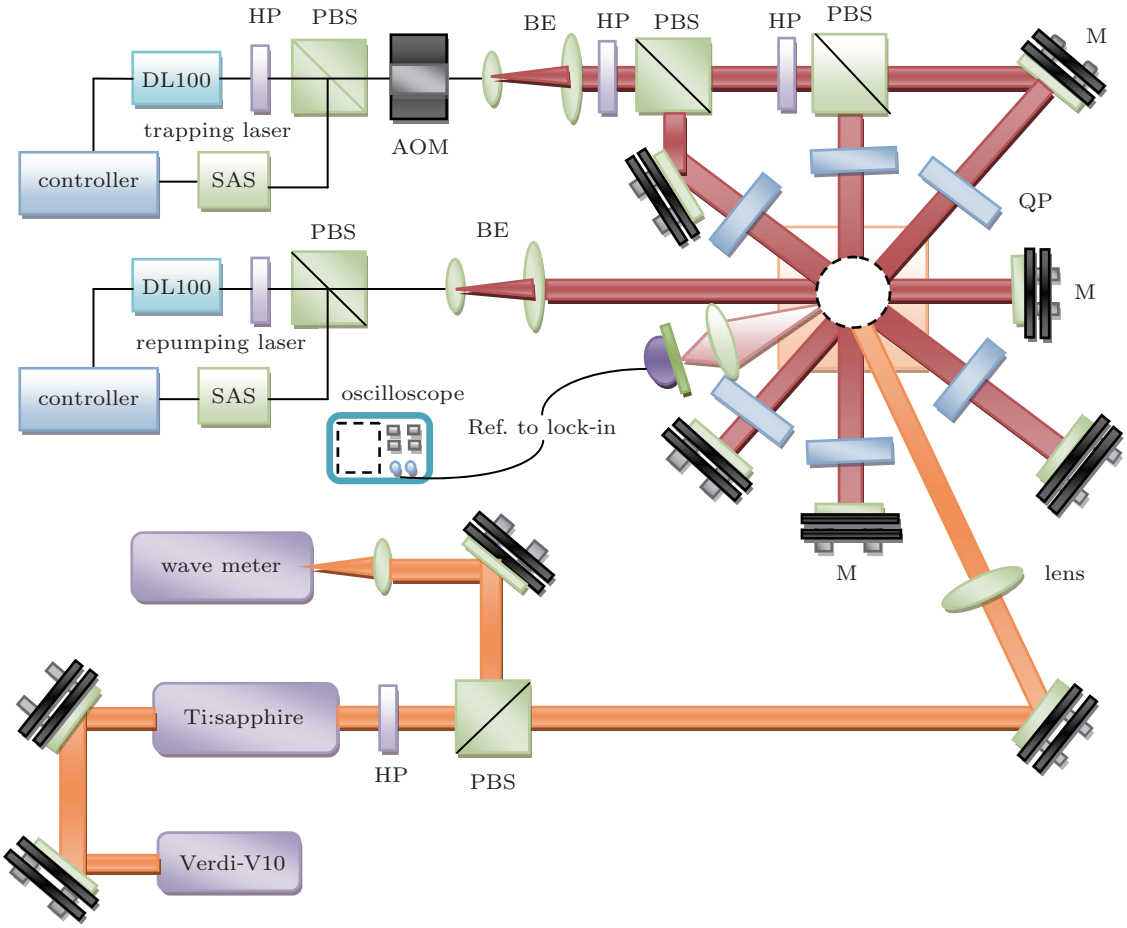


Fig. 1. (color online) The experimental setup. SAS: saturation absorption spectrum; HP: half plate; BE: beam expander; QP: quarter plate; PBS: polarization beam splitter; AOM: acoustic optical modulator; M: reflector.

The PA laser was provided by a widely tunable titanium:sapphire laser system (MBR 110) with a typical power of 600 mW and a line width of about 100 kHz, which was pumped by an argon ion laser. The continuity and linearity of the frequency scans were monitored by a wavelength meter with a high accuracy of 0.002 cm^{-1}. The wavelength meter was repeatedly calibrated against the atomic transition. When the frequency of the PA laser was scanned over the resonant position of the rovibrational levels of molecular excited states, a pair of atoms would absorb a photon and form an excited

molecule. The formed cold excited molecules spontaneously decayed either into two free atoms or into the ground state molecule that we want to obtain. It should be noted that the scanning time should be longer than the atom loading time, which was about 15 s in our experiment.

The fluorescence signal of the ultracold atoms represented the trap-loss of the trapped atoms, so we refer to this technique as trap-loss spectroscopy.[17] The fluorescence signal was collected by a convex lens and detected by an avalanche photodiode with 850 nm band-pass filters. Some of the effective signals submerge into the stray signals, so we used the lock-in method based on modulated ultracold atoms to improve the signal-to-noise ratio of the fluorescence signal. We used a modulation frequency of 3.5 kHz to stabilize the trapping laser frequency. The frequency variation led to the modulation of the intensity of the fluorescence signal, which was derived from the trapping atoms with the same frequency. The modulated signal was demodulated with a lock-in amplifier and recorded with a computer. The trap loss spectroscopy is superior to other techniques, so it can directly provide us with abundant information about the photoassociation of ultracold atoms. In this paper, in order to study the dependence of PA on laser intensity, we only present the trap loss spectrum.

3. Theoretical model

In the regime of low laser intensity, the PA rate is proportional to the laser power, as shown by Prodan *et al.*[12] For the high-intensity regime, a semianalytic model of the PA rate was derived in Bohn's report,[18] in which the scattering process in a dressed molecular state picture was treated. The PA rate coefficient G can be written as an averaged product of relative velocity $v_{\rm r}$, maximal inelastic cross section β, and scattering probability $|S|^2$ as follows:

$$G = \langle \beta v \rangle |S|^2 = \left\langle \frac{\pi}{k^2} v_{\rm r} \right\rangle |S|^2 . \tag{1}$$

According to Eq. (1) the upper limit of the PA rate is the inelastic scattering rate. At a maximum rate, in each scattering event the atom pair absorbs a photon and is converted into a molecule. The scattering probability between the free scattering wave function and a bound molecular state is calculated to have a Lorentzian shape and can be written as

$$|S|^2 = \frac{\gamma \Gamma}{[E-(\Delta+E_1)]^2 + \left(\frac{\gamma+\Gamma}{2}\right)^2}, \tag{2}$$

where Γ is the absorption rate stimulated by the PA laser, γ the decay rate of the bound excited state, Δ the PA laser detuning related to the resonance of rovibrational levels, E the energy difference between the two states, and E_1 the new energy after a light-induced energy shift.

In the theoretical model of Bohn and Julienne[18] for photoassociation in strong laser fields, the stimulated rate of ultracold atoms, Γ, which is dependent on PA laser intensity I, denotes the coupling strength defined by the overlap of the initial continuum wave function $|\Phi(E_{\rm kin} \to 0)\rangle$ with the final bound state wave function $|\Psi(v)\rangle$, and is expressed as

$$\Gamma = 2\pi \left(V_{\rm eg}\right)^2 |\langle \Psi(v) | \Phi(E_{\rm kin} \to 0)\rangle|^2 , \tag{3}$$

where $V_{\rm eg}$ is the Rabi frequency of the electronic transition, which is proportional to the square root of the laser intensity; $I_{\rm s}$ is the saturation intensity introduced according to $I/I_{\rm s} = \Gamma/\gamma$; and γ is the spontaneous decay rate, which is not dependent on PA laser intensity, as it is determined only by the coupling between the bound state wave function Ψ and ultimate free state wavefunction $\Phi_{\rm l}$, and expressed as

$$\gamma = 2\pi |\langle \Psi | \Phi_{\rm l} \rangle|^2 V^2, \tag{4}$$

with V being the artificial coupling element between the bound excited state and the ultimate free state. Γ increases with increasing PA laser intensity I, while γ remains unchanged. When this intensity Γ becomes comparable to the spontaneous emission line width of 12 MHz, the transition begins to saturate, i.e. $\Gamma(I_{\rm s}) = \gamma$, which shows that $I_{\rm s}$ is determined by both Γ and γ, and further determined by the excited state wavefunction Ψ. The excited state wavefunction is different at the different rotational progressions in the same vibrational state, so there is a consequential difference in $I_{\rm s}$. In addition, Γ drops to zero in proportion to $E^{1/2}$, owing to the energy dependence of the Franck–Condon factor in Eq. (3). As this drop-off occurs on an energy scale comparable to γ, it must be included in any PA line-shape analysis, as has been emphasized.

The inelastic-scattering probability in Eq. (2) is in the form of a Lorentzian distribution, but in the low-energy regime both the width Γ and the energy shift E_1 have explicit threshold variations. In order to obtain the maximum amplitude of the resonant line, let $E_{\rm shift}$ be equal to Δ, that is $(-\Delta + E_{\rm shift}) = 0$, $\Gamma/\gamma = I/I_{\rm s}$, where $I_{\rm s}$ represents the saturation intensity of PA. Then the trap loss probability maximum can be written as

$$|S_{\rm max}|^2 = \frac{4\gamma\Gamma}{(\gamma+\Gamma)^2}, \tag{5}$$

and its broadened width is $\delta f_{\rm FWHM} = \gamma + \Gamma$.

Introducing the saturation intensity $I_{\rm s}$ as the intensity where the induced rate Γ is equal to the spontaneous decay rate γ, one obtains a maximal scattering probability

$$|S_{\rm max}|^2 = \frac{4 I I_{\rm s}(J)}{[I + I_{\rm s}(J)]^2}, \tag{6}$$

and a width of

$$\delta f_{\rm FWHM} = \gamma(1 + I/I_{\rm s}). \tag{7}$$

4. Experimental results and discussion

The study of PA spectroscopy is extremely helpful in understanding the underlying dynamics of the PA process for future experiments. The dependence of the PA rate on laser intensity is of particular interest since we can indirectly determine the timescales of molecule formation. The relevant theoretical studies of the PA scattering probability have been reported.[19] The PA rate is predicted to increase linearly with the increase in intensity at low intensities, while various mechanisms responsible for the saturation of the rate at high intensities have been proposed.

The data scans include transitions to 0_g^- and 1_g, as well as 0_u^+ PA resonances.[20] We focus on just the 0_u^+ series in this report. We scan the partial PA spectrum of cesium atoms below the $6S_{1/2}+6P_{1/2}$ limit (See Fig. 2). In this figure, we also present the detailed vibrational spectra of $v = 163$ and $v = 169$ in the inset, respectively. Pichler *et al.*[21] accurately detected the vibrational levels of the 0_u^+ state utilizing the trap-loss spectroscopy technique. The values of vibrational progression v were also assigned from 94 to 220 in Picher *et al.*'s paper. In the present experiment, we adopt their data to determine the vibrational number v in analysis of our data. In order to avoid the disturbances coming from the neighboring levels belonging to the long-range 1_g and 0_g^- states, we concentrate on the transition in the $v = 169$ vibrational state of the long-range 0_u^+ state, induced from free atoms by a PA laser with an absolute frequency of about 11166.445 cm^{-1}.

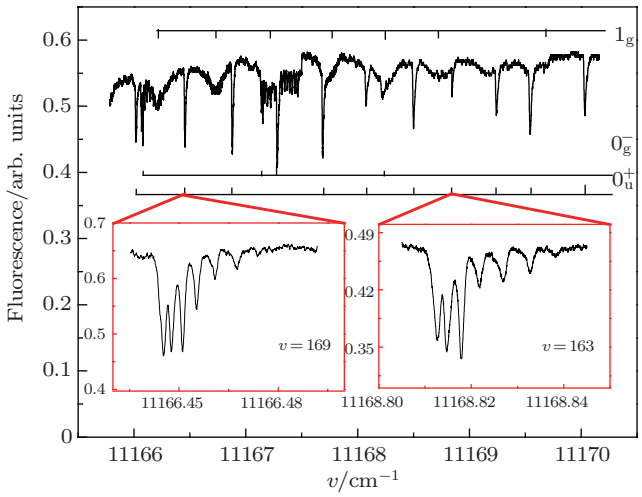


Fig. 2. (color online) Photoassociation below $6S_{1/2}+6P_{1/2}$ for the 1_g, 0_g^-, and 0_u^+ states, with the insets exhibiting the $v = 163$ and $v = 169$ rovibrational structures of the 0_u^+ state.

In order to investigate how and at which intensity the PA rate is saturated, we measure the dependences of PA probability on laser intensity for different rotational progressions in the $v = 169$ vibrational state of the 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ limit. This particular state has a strong PA probability at this position and hence is a good option for our further PA measurements. In Fig. 3, the values of the high-resolution trap-loss probability for four different laser intensities and a scanning speed of 5 MHz/s are shown. The figure clearly shows the rotational structure that is resolved up to $J = 6$ with a high signal-to-noise ratio, and that the peak position remains unchanged. We only show the dependences of PA probability on PA laser intensity for the $J = 1 \sim 6$ rotational levels. Here, $J = 0$ is not considered because its signal amplitude is negligible compared to $J = 1 \sim 6$. In quantitative analysis of the data, a Lorentzian curve is fitted to the rate of each laser intensity, as indicated by the dashed solid line in Fig. 3.

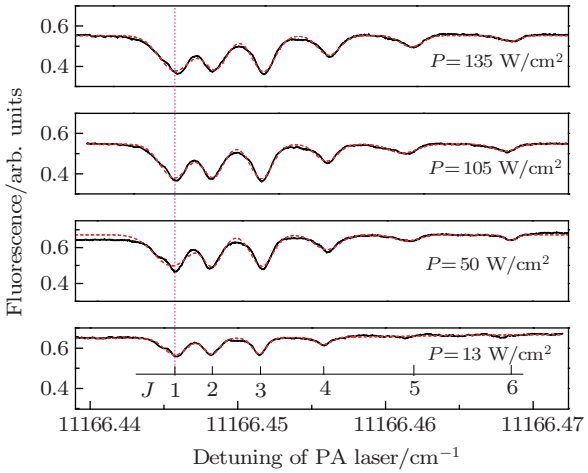


Fig. 3. (color online) The ultrahigh-resolution PA spectra of the $v = 169$ vibrational state for the 0_u^+ long-range state with intensities of 135 W/cm^2, 105 W/cm^2, 50 W/cm^2, and 13 W/cm^2, as shown sequentially from top to bottom. The scanning speed is 5 MHz/s, and in the figure $J = 0$ is inconspicuous. The dashed lines represent Lorentzian fittings to the resonances.

In Fig. 4, the symbols denote the experimental data, while the lines represent the fitting to the saturation model of Bohn and Julienne.[18,22] From the figure, we can easily observe that the PA probability increases with increasing PA intensity, owing to the fact that the photoassociation slows down at large PA intensities. When the PA intensity rises up to 110 W/cm^2, the saturation effect is clearly observable. All the rotational progressions can be fitted by formula (6) within experimental accuracies. We only present the results for $J = 3, 4, 5, 6$, in the figure, respectively. The difference between saturation intensities can clearly be seen for different rotational progressions from the inset of Fig. 4. The error bars indicate the uncertainties for identifying the saturation intensity positions. The saturation intensity of the laser varies between 105 W/cm^2 and nearly 160 W/cm^2, and there is a maximum difference value of about 50 W/cm^2 between the rotational progressions. The difference in saturation intensity between different levels streams from the difference in the transition probability in photoassociation from atoms to molecules between different rovibrational levels. The reason for this is mainly that the Franck–Condon factors of different levels are different. The difference between saturation intensities is not negligible in studying the regime of the excitation of ultracold Cs atoms in the PA process of the 0_u^+ state. The occurrence of the difference between saturation

intensities is due to the intrinsic property of the rovibrational transition of molecules.

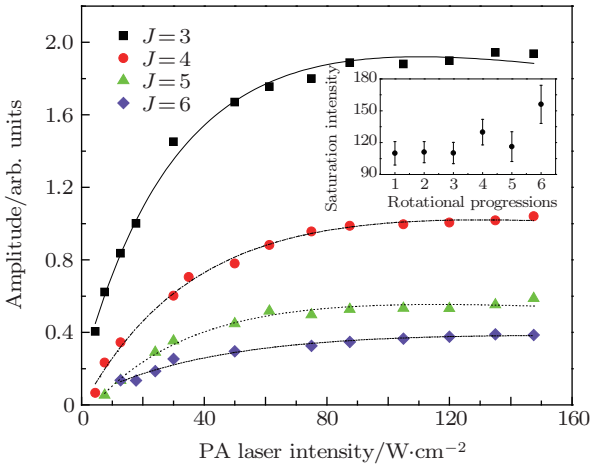


Fig. 4. (color online) Plots of the amplitude of resonant lines for the $J = 3,4,5,6$ rotational levels versus PA laser intensity, with the curves representing the fittings to the saturation model given by Bohn. The inset shows the obtained saturation intensity versus rotational progression.

The line widths of the PA probability[23] curves fitted from Eq. (7) are shown in Fig. 5. Considering possible superposition, we only represent the linear fittings of $J = 1$, 3, and 4. From Fig. 5, a linear dependence of the line width on laser intensity is observed in the whole range of the laser intensity. This can be explained by the model of Bohn and Julienne since the PA rate is still below the unitarity limit. The inset of Fig. 5 shows the slopes of the relevant lines. These line widths[24] will reveal more information about collisional dynamics at ultracold temperatures. Without doubt, these intensity studies reveal additional interesting physics from the PA experiments.[25]

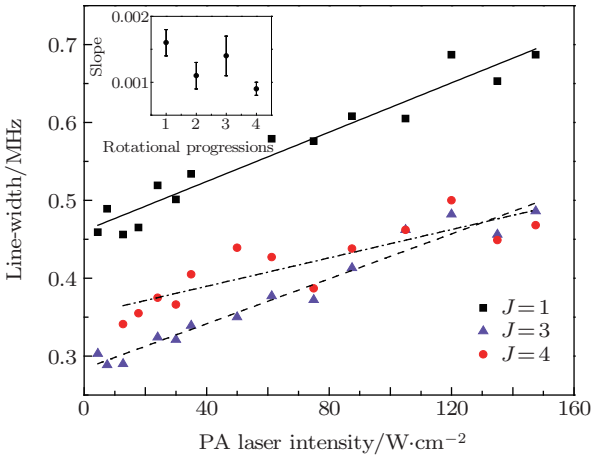


Fig. 5. (color online) The dependences of line width on PA laser intensity for $J = 1$, 3, and 4 of the Cs_2 0_u^+ long-range state. The symbols represent experimental data, while the lines denote the fittings to Eq. (7) as described in the text. The inset shows the slopes of the fitted lines.

The measurements of the intensity dependence of the PA rate of the 0_u^+ long-range molecular state $(6S_{1/2}+6P_{1/2})$ for different rotational levels have not yet been provided by previous reports. The hyperfine rovibrational structure with high-resolution is needed by use of the trap-loss technique in our experiment. The observed high-resolution trap-loss spectrum[21] makes the investigation of the dependences of PA on laser intensity for different rotational progressions possible in our experiment.[18]

5. Conclusions

We obtained the high-resolution trap-loss spectrum by monitoring the fluorescence of trapped cesium atoms as a function of PA laser frequency. The rotational structures of $v = 169$ of the 0_u^+ state $(6S_{1/2}+6P_{1/2})$ are obtained for different laser intensities. In order to study the intensity dependence of the hyperfine resolved spectrum, we quantitatively investigate and observe the dependence of the photoassociation rate on the PA laser intensity of cesium molecules in the 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ limit, and determine the corresponding saturation intensity. Moreover, we analyze the line widths of the resonant lines for different rotational progressions. Both the amplitude and width increases are quantitatively in good consistence with the theoretical model of Bohn and Julienne using the saturation intensity as the only parameter of the model. This work also reveals some interesting features that occur at saturation, and improves the resolution of previous photoassociation experiments. A complete study of the saturation of PA related to the rotational levels is required to understand the PA regime of cesium atoms and to perform the stimulated Raman PA in the well-defined rovibrational levels. To date, this work has not been performed on the saturation effects of the 0_u^+ state below the $6S_{1/2}+6P_{1/2}$ limit, so this study paves the way to investigate these relatively unexplored areas.

References

[1] Thorsheim H R, Weiner J and Julienne P S 1987 *Phys. Rev. Lett.* **58** 2420

[2] Miller J D, Cline R A and Heinzen D J 1993 *Phys. Rev. Lett.* **71** 2204

[3] Zhang W, Xie T, Huang Y, Wang G R and Cong S L 2013 *Chin. Phys. B* **22** 013301

[4] Wang L R, Ji Z H, Yuan J P, Yang Y, Zhao Y T, Ma J, Xiao L T and Jia S T 2012 *Chin. Phys. B* **21** 113402

[5] Zhang H S, Ji Z H, Yuan J P, Zhao Y T, Ma J, Wang L R, Xiao L T and Jia S T 2011 *Chin. Phys. B* **20** 123702

[6] Peter S, Stephan D K, Jorg L, Roland W and Matthias W 2006 *Phys. Rev. Lett.* **96** 023201

[7] Li Y Q, Ma J, Wu J Z, Zhang Y C, Zhao Y T, Wang L R, Xiao L T and Jia S T 2012 *Chin. Phys. B* **21** 043404

[8] Zhang Y C, Wu J Z, Li Y Q, Ma J, Wang L R, Zhao Y T, Xiao L T and Jia S T 2011 *Chin. Phys. B* **20** 123701

[9] Ulmanis J, Deiglmayr J, Repp M, Wester R and Weidemüller M 2012 *Chem. Rev.* **112** 4890

[10] McKenzie C, Denschlag J H, Haffner H, Browaeys A, Araujo E E, Fatemi F K, Jones K M, Simsarian J E, Choa D, Simonib A, Tiesinga E, Julienne P S, Helmerson K, Lett P D, Rolston S L and Phillips W D 2002 *Phys. Rev. Lett.* **88** 120403

[11] Schloder U, Silber C, Deuschle T and Zimmermann C 2002 *Phys. Rev. A* **66** 061403

[12] Prodan I D, Pichler M, Junker M, Hulet R G and Bohn J L 2003 *Phys. Rev. Lett.* **91** 080402

[13] Kraft S D, Mudrich M, Staudt M U, Lange J, Dulieu O, Wester R and Weidemuller M 2005 *Phys. Rev. A* **71** 013417

[14] Mickelson P G 2006 "Saturation Effects in Photoassociation Spectroscopy of ^{86}Sr[D]", Rice University 2006

[15] Hansen D and Hemmerich A 2006 *Phys. Rev. Lett.* **96** 073003

[16] Wang L R, Ma J, Ji W B, Wang G P, Xiao L T and Jia S T 2007 *Laser Phys.* **17** 1171

[17] Ma J, Wang L R, Ji W B, Xiao L T and Jia S T 2007 *Chin. Phys. Lett.* **24** 1904

[18] Bohn J L and Julienne P S 1999 *Phys. Rev. A* **60** 414

[19] Napolitano R, Weiner J, Williams C J and Julienne P S 1994 *Phys. Rev. Lett.* **73** 1352

[20] Ma J, Wang L R, Zhao Y T, Xiao L T and Jia S T 2009 *J. Mol. Spectros.* **255** 106

[21] Pichler M, Chen H M and William C S 2004 *J. Chem. Phys.* **121** 1796

[22] Bohn J L and Julienne P S 1996 *Phys. Rev. A* **54** 4637

[23] Simoni A, Julienne P S, Tiesinga E and Williams C J 2002 *Phys. Rev. A* **66** 063406

[24] Wu J Z, Ma J, Ji Z H, Zhang Y C, Li Y Q, Wang L R, Zhao Y T, Xiao L T and Jia S T 2012 *Chin. Phys. B* **21** 093701

[25] Gomez E, Black A T, Turner L D, Tiesinga E and Lett P D 2007 *Phys. Rev. A* **75** 013420