High-resolution photoassociation spectroscopy of ultracold Cs_2 long-range 0_g^- state: The external well potential depth*

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High-resolution photoassociation spectroscopy is reported using a modulation spectroscopy technology in a cesium atomic magneto-optical trap. The two lowest vibrational levels have been experimentally observed which have been theoretically predicted in [*Phys. Rev.* A **75** 052501 (2007)]. A new potential curve is obtained by using the Rydberg–Klein–Ress method with a well depth of -82.384 ± 0.026 cm^{-1}, which is deeper than the result of previous experiment (-77.909 cm^{-1}) and the theoretical prediction (-81.6445 cm^{-1}).

Keywords: ultracold molecules, photoassociation, trap-loss spectroscopy, potential depth

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1. Introduction

Recently, great attention has focused on the ultracold molecule due to their wide applications in ultracold chemistry,[1] quantum computing,[2] and molecular optics.[3,4] Photoassociation (PA), taking advantage of the technology of laser cooling and trapping atoms, opens a convenient and robust way to obtain ultracold molecules.[5,6] In the photoassociation process, two colliding ultracold ground-state ultracold atoms resonantly absorb a photon and form an electronically excited molecule, which populates in a certain rovibrational level with large internuclear separation and is usually called long-range molecule. A high-resolution PA spectroscopy[7] can provide information on the molecular levels in the excited state. The spectroscopy data is essential to determine the molecular parameters[8–12] and characterize the potential curves.[13]

For PA experiments of homonuclear alkali species, such as the cesium and rubidium experiments, the double-well structure of the long-range 0_g^- molecular state correlated to the dissociation limit is an important and interesting feature, as the external well can be chosen as a suitable intermediate step in the course of forming ground-state molecules in their absolute ground state.[14] The external well of the 0_g^- double-well curve in alkali dimmers belonging to pure long-range molecule states, as described by Stwalley *et al.*[15] is almost entirely located within the asymptotic region, where the chemical exchange energy contribution is weak enough to be treated as a perturbation. With highly sensitive detection methods,[16,17] research on pure long-range molecule has achieved significant progress in applications and precise measurements, such as the measurement of long-range rovibrational molecule coefficients,[18,19] absolute frequency stabilization of a diode laser,[20] and resonant four-body interaction in cold cesium Rydberg atoms.[21] The pure long-range 0_g^- state of Cs_2 PA spectroscopy was first obtained by Pillet's group using ionization detection technology in 1999,[12] where they labeled the lowest levels as $v = 0$. The external well potential was obtained with the spectroscopy data and the potential absolute well depth is 77.94 ± 0.01 cm^{-1} from the dissociation limit (11732.183 cm^{-1}). However, according to the theoretical calculation,[22] in which the external well of the 0_g^- state Cs_2 potential curve has been precisely analyzed, there are still other two levels populated at the energy position lower than $v = 0$ obtained by Pillet *et al.* In the theoretical calculation of Ref. [22], the values of the radiative lifetime and the van der Waals coefficient C_7 of the ground-state cesium molecule and other parameters are modified with the two levels. If the prediction is true, the potential curves based on the former vibrational levels need to be modified and the potential depth can be deeper in view of the new lowest levels.

In this paper, the ultracold cesium molecular spectroscopy is obtained through PA technology. Two new lowest levels of the Cs_2 pure long-range 0_g^- state is observed by high-resolution PA spectroscopy. A modified potential curve near the equilibrium position is obtained through the Rydberg–Klein–Ress method. In addition, a new potential depth is obtained.

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2. Experiment setup

The experiment setup is schematically depicted in Fig. 1. About 10^7 ^{133}Cs atoms are captured in a conventional vapor loaded MOT,[23] yielding an atomic density of 10^{12} cm^{-3} deduced by the absorption method.[24] The temperature of atom samples is adjusted to be about 200 μK by using the time-of-flight method. The trapping laser and the repumping laser are all frequency-stabilized diode laser (Toptica DL 100). The trapping laser is locked to two natural linewidths which are red detuned from the Cs atomic transition $6S_{1/2}(F = 4) \longrightarrow 6S_{3/2}(F' = 5)$ by using saturated absorption spectroscopy. The repumping laser is locked to the $6S_{1/2}(F = 3) \longrightarrow 6S_{3/2}(F' = 4)$ atomic transition.

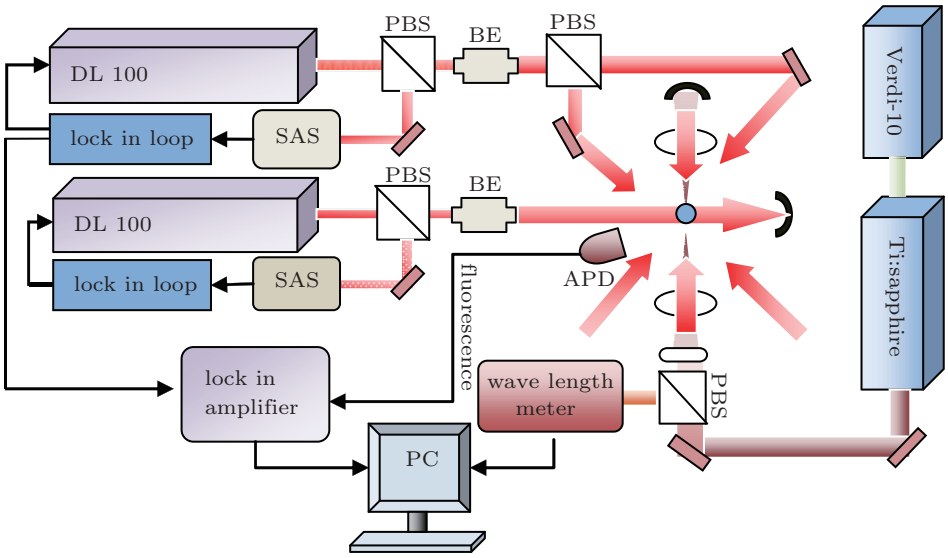


Fig. 1. (color online) Experiment setup. PBS: polarization beam splitter; BE: beam expander; SAS: saturated absorption spectrum; PC: personal computer.

A widely tunable Ti:sapphire laser (Coherent MRB 110) system with a linewidth of about 110 kHz and a typical out power of 900 mW serves as the photoassociation laser source. The PA laser beam is focused with a waist of 500 μm to cover the atom cloud in MOT. As the PA laser scans, the frequency is measured by a wavelength meter (High Finesse-Angstrom WS/7R) with an accuracy of 0.002 cm^{-1}. The PA process is monitored though the trap loss spectroscopy technique.[25] It is realized by recording the trap fluorescence, which is proportional to the atom number. The fluorescence formed by the trap atoms is collected by convex lens and detected by an avalanche photodiode (APD Hamamatsu S3884) with 850-nm band-pass filters.

In the experiment, the direct recording of the fluorescence signal seems impossible, as the signal is so weak that it is submerged in the electronic noises. In order to improve the signal-to-noise ratio, we use the lock-in method, which is base on the modulating fluorescence of ultracold atoms.[26] The modulated fluorescence signal was demodulated through a lock-in amplifier (SR830) with an optimized integration time of 300 ms.

3. High sensitive rovibrational spectroscopy

The PA laser frequency is tuned far from the near-dissociation limit up to -83 cm^{-1} according to the theoretical prediction. The scan rate is set as small as 0.3 MHz/s with power of 800 mW.

Figure 2 shows typical PA spectroscopy for the lowest two vibrational states of the electronic 0_g^- state, whose detuning with respect to the $6P_{3/2}$ dissociation limit is ~ -80 cm^{-1} and ~ -82 cm^{-1}, respectively. The maximum signal-to-noise ratio of the spectral signal detected by the modulation spectroscopy technology spectroscopy technique is up to 14.5. The rotational progression is observed up to $j = 4$ for $v = 1$ in this way. However, the cases of ($v = 0$, $j = 0$) and ($v = 0$, $v = 1$) are not observed mainly due to the small Franck–Condon factors. The small transition probabilities in photoassociation from atoms to molecules in these levels result in small losses of atoms. The two spectra show similar intensity undulation as reported in Ref. [17], which are due to the variation in the Franck–Condon factors for the photoassociation transitions between the initial state of the two cold colliding atoms and the final rovibrational levels of the long-range states.[27]

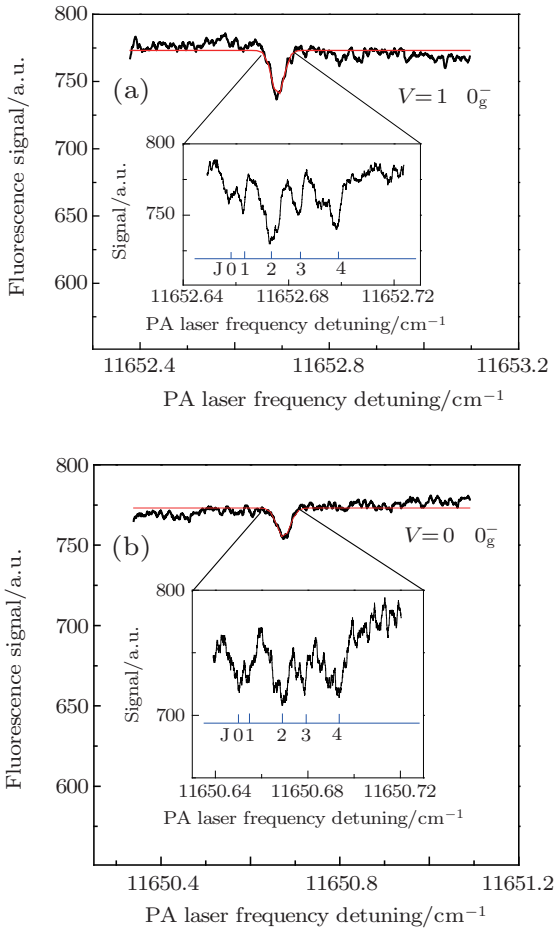


Fig. 2. (color online) (a) The $v = 1$ vibration position of the pure long-range 0_g^- state. (b) The $v = 0$ vibration position of the pure long-range 0_g^- state. The inset shows the rotational progressions for each vibration state.

4. Examination of the external well potential depth

The quantitative analysis of the energy position of long range molecular states are often performed by fitting procedures using the LeRoy–Bernstein formula[28] (LRB) or the Rydberg–klein–Ress (RKR) method.[29] The LRB method is usually feasible in the near dissociation limit range. We use the RKR method with the Dunham procedure, which is adapted to the short-range exponential behavior of most molecular potentials. The turning points are determined by the Klein integrals as follows:

$$R_+(v) - R_-(v) = \frac{2\hbar}{\sqrt{2\mu}} \int_{v_{\min}}^{v} \frac{\mathrm{d}v'}{[G(v) - G(v')]^{-1/2}}, \quad (1)$$

$$\frac{1}{R_-(v)} - \frac{1}{R_+(v)} = \frac{\hbar}{2\pi\sqrt{2\mu}} \int_{v_{\min}}^{v} \frac{B(v)\,\mathrm{d}v'}{[G(v) - G(v')]^{-1/2}}, \quad (2)$$

where the $R_-(v)$ and the $R_+(v)$ are the inner and the outer turning point corresponding to energy $G(v)$, the lower bound $v_{\min}$ is the value of vibration number at the potential minimum (equal to -0.5), μ is the reduced mass of ^{133}Cs, and $B(v)$ is the rotational constant of each vibration level. In order to solve the integral, we take advantage of the Dunham procedure,[30] the energy $E(v, j)$ belonging to a single electronic state of a diatomic molecule is written as

$$E(v, j) = \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} Y_{lm} (v + 1/2)^l [j(j+1)]^m. \quad (3)$$

The Y_{lm} term is known as the Dunham coefficient. This expansion can also be written as

$$E(v, j) = \sum_{m=0}^{\infty} K_m(v) [j(j+1)]^m, \quad (4)$$

$$K_m(v) = \sum_{l=0}^{\infty} Y_{lm} (v + 1/2)^l. \quad (5)$$

In this expansion, $K_0(v)$ is identified as the rotationless vibrational energy $G(v)$ and the $m = 1$ terms $K_1(v)$ as the inertial rotational constant $B(v)$. The constants $K_2(v) = -D(v)$, $K_3(v) = H(v)$, ..., are the centrifugal distortion constants. Dunham coefficient $Y_{01} = B_e$ is regarded as the harmonic and rotational constant at equilibrium.

Table 1. The primary molecular parameters of the 0_g^- state of Cs_2 obtained from the spectrum together with the results of Refs. [13] and [22]. All qualities are in units of cm^{-1} except R_e and $a_0 = 0.5291772$ Å.

	This work	1999[13]	Theory[22]
Y_{10}	2.1883	1.7855	–
Y_{20}	-0.5583×10^{-1}	-0.1385×10^{-1}	–
Y_{30}	0.2173×10^{-4}	0.2662×10^{-4}	–
Y_{40}	-3.9815×10^{-5}	–	–
Y_{01}	0.1567×10^{-2}	0.1667×10^{-2}	–
Y_{11}	-0.2116×10^{-4}	-0.1576×10^{-4}	–
R_e	$24.07a_0$	$23.36a_0$	$23.453a_0$

A Dunham fit, according to Eq. (3), of the experimental data shown in Fig. 2 gives the Dunham coefficients quoted in Table 1. The R_e is obtained from the relational of $Y_{01} = B_e = \hbar/8\pi^2 c\mu R_e^2$, where c is the speed of light and μ is the reduced mass of ^{133}Cs. The R_e in this work is longer than the theoretical prediction and the result of Pillet's group in 1999. The major part of the rotational constant $B(0) = Y_{01}$ is smaller than the result of 0.0016 cm^{-1}. The lowest levels ($v = 0$, $j = 0$; $v = 0$, $j = 1$) cannot be read out directly from the PA spectroscopy. However, the lowest level ($v = 0$, $j = 0$) is important to determine the potential depth. We use the rotational constant method to calculate the levels. The long-range interaction between two constituent atoms of the excited molecules formed by photoassociation is weak, compared with ordinary chemical bond, so the rotation model can be written as[31]

$$\frac{E}{hc} = Bj(j+1) - Dj^2(j(j+1)), \quad (6)$$

where E is the rotational eigenvalue, h is Planck's constant, c is the light speed, B is the rotational constant, D is the centrifugal distortion constant (both B and D are in units of cm^{-1}),[32] and j is the rotational quantum number.

For the same vibrational quantum number, the interval ΔE between two neighboring rotational levels is

$$\Delta E = [E(j+1) - E(j)]/hc = 2B(j+1) - 4D(j+1)^3. \quad (7)$$

The rotational constant B and centrifugal distortion constant D can be obtained by fitting experimental data into Eq. (6). From the Dunham fitting, $B(0) = Y_{01} = 0.001567$ cm^{-1}, $D(0)$ is several orders smaller than $B(0)$, so the effect of $D(v)$ can be ignored. Finally, the energy for (v = 0, $j = 0$) and ($v = 0$, $j = 1$) is 11650.6600 cm^{-1} and 11650.6570 cm^{-1}. The interval is larger than the accuracy of our wavelength meter (0.002 cm^{-1}).

The coefficients from Table 1 are taken into the RKR calculation. Finally, the RKR turning points up to the last observed vibrational level $v = 18$ and corresponding energy $G(v)$ and the rotational constant $B(v)$ are obtained. The Cs_2 0_g^- state potential curve is plotted in Fig. 2, in which the potential minimum value together with the two lowest levels (tawny and orange lines) is -82.384 ± 0.06 cm^{-1}. We can find that the red curve in Fig. 2 is 77.94 ± 0.01 cm^{-1},[13] and the theoretical prediction in Ref. [22] is -81.6445 cm^{-1}. Compared with previous results in Ref. [13], the present result for the modified potential curve is 4.44 cm^{-1} deeper due to the added two new levels. In the case of theoretical prediction, it is $0.73 + 0.06$ cm^{-1}, and the accuracy is 0.91%, which is suitable for the new result. The potential minimum is $24.07a_0$, a little longer than $23.453a_0$ reported in Ref. [22].

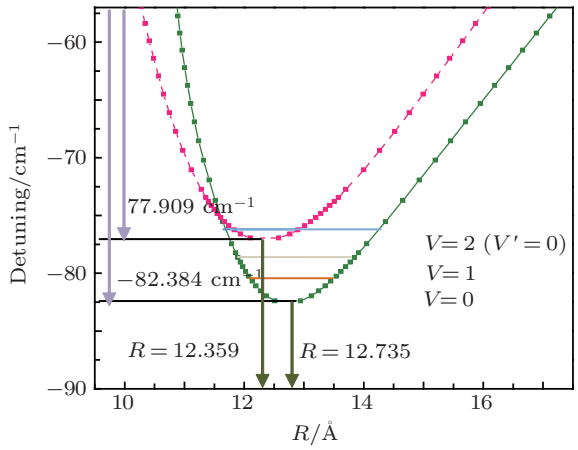


Fig. 3. (color online) The Cs_2 0_g^- state potential curve of the external well. Green dotted lines: the potential curve with the lowest levels. Red dotted lines: the part of the potential in Ref. [13]. The experimental detuning δ^{exp} was deduced from the relation of $\delta^{exp}(v) = \delta^{exp}(0) + G(v) - G(0)$, where $G(0) = 1.03$ cm^{-1}, a little larger than 0.89 cm^{-1}.[13] $G(v) = -81.5217$ cm^{-1}, which is the detune of the lowest levels ($v = 0$, $j = 0$) to $6P_{3/2}$ dissociation limit (11732.176 cm^{-1}). The tawny and orange line represent the new two levels. While, the blue line, which has been regard as $v = 0$, is labeled as $v = 2$ level.

5. Conclusion

In conclusion, we observe the lowest molecular vibrational levels for Cs_2 0_g^- state as predicted. With these levels, we obtain a deeper potential curve at the equilibrium position and the external well of the depth is -82.384 ± 0.06 cm^{-1} with the potential minimum being $24.07a_0$. As the method of RKR is especially suitable for the analyses of lower levels, the turning point is credible, the curve of this range is successful, and it is enough to find out the depth of the external well.

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