Precise measurement of the line width of the photoassociation spectra of ultracold molecules by a frequency shifter*

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We propose a technique to precisely measure the line width of the photoassociation spectra of the excited cesium molecule by using a frequency shifter to generate two laser beams with a precise frequency difference. A series of photoassociation spectra are recorded with two laser beams induced molecular lines, whose peak separation serves as an accurate frequency ruler to measure the line width of the photoassociation (PA) spectra. The full width half maximum line width was studied as a function of PA laser intensity. The extrapolated value at zero laser intensity is (34.84 ± 0.22) MHz. By analyzing other broadening mechanisms, a value of (32.02 ± 0.70) MHz was deduced. It is shown that this scheme is inexpensive, simple, robust, and is promising for applications in a variety of other atomic species.

Keywords: laser cooling, molecular spectroscopy, line shapes and width

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1. Introduction

The great advancement of the experimental techniques of laser cooling and trapping of neutral atoms has led to the production of quantum degenerate atomic Bose and Fermi gases.[1] The extension of cooling and preparing molecules into their rovibrational ground state will open up the possibilities to precisely manipulate the internal and external degrees of freedom of the molecules. It is expected that ultracold molecules can be used to obtain high-resolution spectroscopy and measure the fundamental constants,[2,3] investigate ultracold collisions and chemistry,[4] develop quantum computation techniques,[5] and produce molecular Bose–Einstein condensates (BEC).[6] However, the lack of closed two-level optical pumping scheme for recycling population in molecular structure precludes the direct cooling of molecules.[7] Ultracold molecules in and below the μK–nK range could only be produced from cold atoms via association techniques. Recently, association via Feshbach resonances has directly produced quantum degenerate gas but only in very weakly bound states.[8] Moreover, these molecules are generally unstable if they are composed of bosonic species. An alternative standard and common approach to producing more stable (deeply-bound) molecules is photoassociation (PA).[9] In the PA process, two free cold ground state atoms resonantly absorb one photon and produce an electronically excited molecule in a given ro-vibrational state with a large internuclear separation, usually called a long-range molecule. PA offers a unique opportunity to study the long-range interaction between atoms, which plays an important role in understand-

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ing the weakly bound excited state molecules and cold collisions.[9] Up to now, PA of cold atoms has been demonstrated for all alkali atoms even in alkaline earth atomic species.[9−14] Recently, argon atomic photoassociations have been reported.[15]

The PA spectroscopy of trapped alkaline atoms has become a dynamic field of enthusiastic research recently. A deep understanding of the line shape is crucial in using photoassociation spectroscopy to obtain ground state scattering lengths for Bose–Einstein condensate studies. On the other hand, line-width measurements are fundamental for understanding the atomic structure. The atomic structure theory can be testified by comparing the precisely measured quantities of transition energies and excited state lifetimes with that obtained from the calculated energies and transition matrix elements. A number of experimental studies about line width have been performed. By probing the atomic hyperfine structure, the natural Lorentzian linewidth of Na $3^2S_{1/2} \rightarrow 3^2P_{3/2}$ transition was precisely measured, upon which the atomic lifetime was determined accurately.[16] Line shape as well as line shifts were found to be a function of intensity and detuning in two-photon photoassociation spectra in preparing of cesium ground state molecules in a Λ-type scheme.[17] These studies have shown the intensity dependence for a chosen rotational level in a vibrational state. However, a systematic measurement of the intensity dependence of the PA line width for the rotational levels in a certain vibrational state has not been performed, which may allow to analyze the molecular bound state wavefunction.[1] The investigation of the dependence of PA resonant line width on the laser intensity is of importance due to its role in understanding the mechanism of resonant excitation of a pair of free atoms into molecule residing in excited state. The involved dependence of line width of resonant signal on PA laser intensity offers a direct and effective indication for PA rate.

2. Experiment

The experiment was carried out in a conventional cesium vapor cell magneto-optical trap (MOT).[18,19]

The configuration of the setup is shown schematically in Fig. 1. Three pairs of mutually orthogonal laser beams comprise the MOT with opposite circular polarizations and intensities of about 3 mW/cm^2. A typical magnetic gradient of 15 G/cm for the MOT was generated by a pair of anti-Helmholtz coils. The trapping laser frequency was red detuned from the $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$ atomic transition for about two natural line-width. The repumping laser was locked to the $6S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ transition. Thus, a cold cesium atomic ensample was prepared with a number of $\sim 10^7$ and a peak density of $\sim 10^{11}$ cm^{-3}. The temperature of the cold atomic cloud was about 100 μK by time-of-flight method. Trapping laser as well as repumping laser (Toptica, DL100, ∼ 160 mW, line-width 0.8 MHz) was frequency-stabilized using standard absorption technology. The fluorescence yielding by the cold atomic cloud is collected by a convex lens and detected by a silica avalanche photodiode (APD Hamamatsu S3884) with 850 nm band-pass filters.

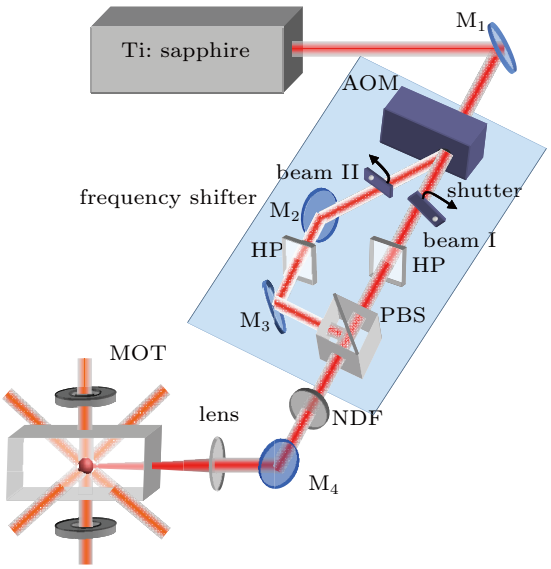


Fig. 1. (colour online) Schematic diagram of the experimental setup. The red line is the PA laser beam path while the orange line is the MOT beam path. NDF: neutral density filters; HP: half wave plate.

Photoassociation (PA) was accomplished by a continuous wave (CW) widely tunable Ti: sapphire laser system (Coherent MBR110, typical line-width < 100 kHz) pumped by a diode laser (Verdi-10). The PA laser frequency was measured by a wave meter (HighFinesse-Angstrom WS/7R) with an accuracy of 60 MHz. The meter was repeatedly corrected against the cesium atomic hyperfine resonance transition, $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$. A frequency shifter, as shown highlighted in Fig. 1 in a trapezoid configuration, is mainly composed of an Acousto-Optical Modulator (MT110-B50A1-IR, AA Optoelectronic). The AOM diffracts the incident PA laser

beam into different orders (1st, -1st$\cdots$), frequencies of which are shifted from that of the PA laser the (the 0th order after transmitting the AOM) by an interval of 85–135 MHz. This frequency interval, which is dependent on the oscillator of the AOM, can be changed through the offset potentiometer of the driver. The Brag angle between the incident beam and the acoustic wave was adjusted to make sure that both the intensities of the 0th (beam I) and the -1st order (beam II) are the same and make up the major proportion ($\sim 95\%$) of the input laser intensity. The half wave plates and polarization beamsplitter (PBS) were used to superimpose the two beams into the same alignment and control the intensities.

The typical output of the PA laser is nearly 900 mW with pump power of 9.5 W. After transmitting the AOM, beam I as well as beam II was focused to a diameter of 500 μm, resulting in an available intensity of nearly 250 W/cm^2. As the PA laser frequency scans over the resonant position of the excited Cs molecular state, spontaneous emission of the excited molecules leads to formation of ground state molecules or dissociation into two free atoms, both of which result in depletion of the atomic population from the MOT. Thus, the analysis of the trap losses is a convenient way to detect PA. The PA trap-loss experiments with cold cesium atoms have enabled the spectroscopy of the three attractive Hund's case (c) states populated below the $6S_{1/2}+6P_{3/2}$ dissociation limit: 0_g^-, 0_u^+, and 1_g. The lock-in detection technique via modulating fluorescence of cold atoms was utilized to improve the sensitivity and signal-to-noise ratio of detection. A modulation frequency of 2.2 kHz was chosen to lock the MOT laser frequency. The modulated fluorescence was demodulated by a lock-in amplifier (SR830). PA spectra were thus obtained through recording the fluorescence as a function of PA laser frequency. High sensitivity and high resolution of the trap loss spectroscopy for long-range molecular 0_g^- state permits further research on molecular constants.[20−22]

3. Experimental results and discussion

This experiment shows how the photoassociation transition line-width can be determined by the technique using the frequency shifter. A series of photoassociation spectra were firstly acquired when the atomic sample was alternately illuminated by the PA beams. As a pure long-range state, the 0_g^- state possesses a double-well structure, which provides levels lying in the external well for efficient PA and substantial decay to the levels of the lowest triplet electronic ground state.[23] For example, for $\nu = 17$ of Cs_2 0_g^- pure long-range state, whose detuning compared with the $6P_{3/2}$ dissociation limit is ~ 51 cm^{-1}, is used to study the line-width. The peak interval between rotational levels $J = 2$ and $J = 3$ for vibrational level $\nu = 17$ is about 242 MHz, while that for $J = 3$ and $J = 4$ is 342 MHz, for $J = 4$ and $J = 5$ is 411 MHz. Figure 2 shows a typical high resolution PA spectrum for this state, which specifies rotational progressions of from $J = 0$ to $J = 5$. Due to the relatively low temperature of the atomic cloud, additional angular momentum (orbital angular moment due to p-wave or d-wave resonance) makes no contribution. Thus, molecules on large rotational levels $J > 6$ are not considerably produced. The maximum signal-to-noise ratio of spectral signal detected by trap-loss spectroscopy technique is up to 48, while the loss ratio of the signal is up to 56%. The Ti-Sapphire laser was scanned by 2 GHz over 400 s, which was unchanged during the experiment. The linearity and continuity of the scanning were monitored through the wave meter. In case of laser mode-hopping, or a bad scanning, the scan was repeated.

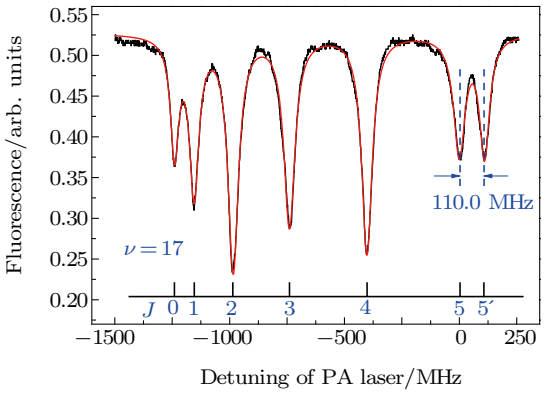


Fig. 2. (colour online) Rotational resolved trap-loss spectrum for $\nu = 17$ of Cs_2 0_g^- pure long range state. Rotational progressions from $J = 0$ to $J = 5$ induced by beam I are indicated with a high SNR. $J = 5'$ induced by beam II is combined in the spectrum. A peak separation between $J = 5$ and $J = 5'$ of 110 MHz is utilized as an optical ruler to measure the Photoassociation line-width. The red curve is the corresponding Lorenzian multi-peaks fitting result.

By alternately illuminating the magneto-optical trap with the two PA laser beams shifted with a fixed frequency amount from each other, a photoassociation spectrum was recorded in which either beam gives rise to a molecular line. At the beginning of an experimental cycle, beam I shines the cold atomic cloud and induces photoassociation while beam II is blocked by a shutter. Ro-vibrational progressions from $J = 0$ to $J = 5$ appear one after another and form a trap loss spectrum as beam I scans over 11681.32 cm^{-1}, corresponding to the vibrational level of $\nu = 17$ in the Cs_2 0_g^- pure long-range state. The intensity undulation of the molecular lines in the spectrum stems from the fluctuation of the Franck–Condon factors for the PA transitions between the initial state of two cold free atoms and the final ro-vibrational levels of the 0_g^- state. Beam I is shielded quickly once the $J = 5$ resonance peak is nearly fully scanned over. Simultaneously, the shutter of the beam II is opened and the atom cloud is illuminated in succession by beam II, which is the -1 order diffracted by the AOM from the input PA laser beam. Thus, beam II induces a $J = 5$ resonance line again in the spectrum (for denotation, $5'$ in Fig. 2). Either of the two beams is rapidly shuttered or shone, so the independent resonance peaks of the same rotational progression are clearly demonstrated in the spectrum.

The line-width broadening, loss ratio and the frequency shift of a specific PA rotational progression induced by beam I equal that of the corresponding level induced by beam II, under the condition that both the intensities of the two beams are the same. Consequently, as shown in Fig. 2, the frequency separation between the two peak positions of $J = 5$ and $J = 5'$ is exactly the frequency difference of the two laser beams, i.e., the frequency of oscillator of the AOM with an accuracy of kHz, which serves as a precise optical ruler. For convenience and to avoid disturbance of the neighboring rotational levels, the central frequency 110.0 MHz of the AOM is taken as the frequency difference and set fixed by the offset potentiometer in the AOM driver. All the photoassociation line-widths of the rotational progressions ($J = 0$–5) on the same vibrational level are directly read out according to this ruler, whose scale is 110 MHz. By increasing or decreasing the PA laser intensity using different natural density filters, the photoassociation line-widths will broaden or narrow accordingly.[24] In order to obtain the best result, we restricted the experimental data to the rotational level $J = 2$, corresponding to the most intense line of the observed rotational progressions in $J = 0$–6. The fitted full width at half maximum (FWHM) line-width was plotted versus the PA laser intensity, as shown in Fig. 3. The line-width at zero laser intensity was determined by extrapolating the fitted line, with a value of 34.84 MHz.

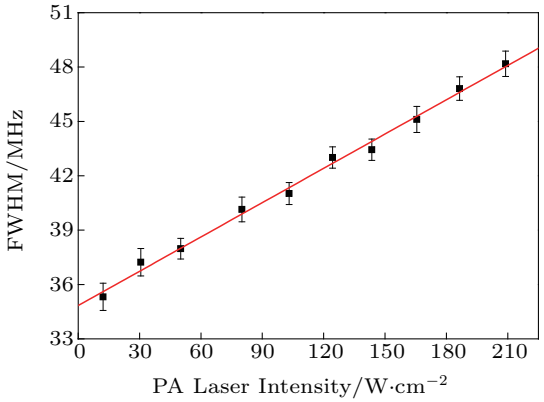


Fig. 3. (colour online) Dependence of the observed FWHM line-width on the photoassociation laser intensity. The straight line was fit to measured values to extrapolate the line-width at zero intensity.

All the mechanisms that could contribute to the line-width are listed in Table 1. The largest broadening effect stems from the frequency drift of the trapping laser, which is due to the accuracy of the frequency locking electronics and the laser line-width which is less than 0.5 MHz within 5 μs. The trapping laser frequency was stabilized by saturation spectroscopy technology which provides an accuracy of 0.81 MHz by long term observation over 1 h. Other possible broadening mechanisms are also estimated.[25] Because the density of ultracold atoms was several orders of magnitude lower than that of the Bose–Einstein condensates or other quantum gases, the collisions in the cold atomic sample were trivial. On the other hand, under the background pressure of 10^{-7} Torr, the number and density dependent effects such as the background and cold atom collisions play a negligible role in determining the line shapes. The residual Doppler broadening was evaluated via Voigt profile calculation with a correction of 8 kHz. The scheme we proposed is independent of the line-width of the PA laser. However, the power broadening is estimated to be about 2 MHz. Thus, the final result of the PA line-width is (32.02 ± 0.70) MHz. Compared with the twice of the cesium atomic natural linewidth of $5.234 \times 2 = 10.47$ MHz,[26] the value we acquired

is comparatively large. We estimate that the temperature, the phase density and the vacuum degree lead to the broadening of the photoassociation linewidth.

Table 1. Determination of photoassociation linewidth.

Mechanism	Value/MHz
extrapolated line-width at 0 intensity	34.84 ± 0.22
Frequency drifts of trapping laser	-0.81 ± 0.01
Collisional broadening	< 0.01
Residual doppler broadening	-0.01
Power broadening	$\sim -2 \pm 0.5$
Final result for PA line-width	32.2 ± 0.70

4. Conclusion

In conclusion, a frequency shifter has been proposed for deflecting a laser beam to investigate the photoassociation line-width of excited cesium molecules. The measurement sensitivity we achieved is high enough to carry out further research, for example, on the scattering length for cesium atomic condensate. A linear dependence of the line-width on the photoassociation laser intensity is found. Different line-width broadening schemes are analyzed and taken into account for the determination of the PA line-width. The scheme we proposed has several advantages. First, the photoassociation line-width is directly measured in a novel way, which is not influenced by the laser intensity induced frequency shift of the rovibrational energy levels. Furthermore, a high quality cavity, which is rather difficult to adjust and expensive, is not required. Finally, the simple and robust scheme should work for a number of other atomic species.

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