Cold cesium molecules produced directly in a magneto–optical trap*

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We report on the observation of ultracold ground electric-state cesium molecules produced directly in a magneto–optical trap with a good signal-to-noise ratio. These molecules arise from the photoassociation of magneto–optical trap lasers and they are detected by resonantly enhanced multiphoton ionization technology. The production rate of ultracold cesium molecules is up to 4×10^4 s^{-1}. We measure the characteristic time of the ground electric-state cesium molecules generated in the experiment and investigate the Cs_2^+ molecular ion intensity as a function of the trapping laser intensity and the ionization pulse laser energy. We conclude that the production of cold cesium molecules may be enhanced by using appropriate experimental parameters, which is useful for future experiments involving the production and trapping of ultracold ground electric-state molecules.

Keywords: photoassociation, ultracold molecules, multiphoton ionization

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1. Introduction

Over the past 20 years, the field of ultracold atoms physics has made enormous accomplishment with the development of the laser cooling and trapping of neutral atoms,[1] such as the achievement of atomic Bose–Einstein condensation.[2] Similar advances are expected in the field of ultracold molecules, for example, new platforms for quantum computing, precise control of molecular dynamics, molecular clocks, tests on fundamental physical constants, nanolithography and Bose-enhanced chemistry.[3] The direct application of laser cooling and trapping techniques to ultracold molecules is very difficult because of the complex levels and lack of closed two-level transitions. Some schemes, including buffer gas cooling,[4] Stark deceleration,[5] and velocity selection,[6] and inelastic scattering in crossed molecular beams,[7] have produced and trapped cold molecules successfully without using laser cooling techniques. However, the only way to produce molecules in the sub-milliKelvin temperature range is the association of ultracold and relatively dense atoms, for example, magnetoassociation (MA) and photoassociation (PA). MA makes use of Feshbach resonances to couple an open collisional channel to a closed molecular one, converting a fraction of the atoms into dimers.[8] A major breakthrough using the MA technique was the observation of molecular Bose–Einstein condensates.[9,10] PA, a well-known efficient process for the formation of ultracold molecules with a rate as high as $\sim10^6$ s^{-1}–10^7 s^{-1}, starts from a pair of ultracold atoms absorbing a photon to form an ultracold molecule in a short-lived excited electronic state.[11] For instance, ultracold ground state K_2 molecules have been produced at the lowest vibrational level $v = 0$ via a two-step PA scheme.[12] A sequence of a PA step followed by an absorption–emission transfer has created ultracold ground state RbCs molecules in $v = 0$.[13] Up to now, the PA technique has been used to form many alkali dimers, such as Cs_2,[14] Na_2,[15] and Rb_2,[16] or alkaline earth metals, Ca_2[17] as an example, and other heteronuclear

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species with two different alkali dimmers, for instance, KRb,[18] NaCs,[19] and LiCs.[20]

In the research process of PA techniques, some researchers[14,21] have found that Cs_2 molecules and Rb_2 molecules can be observed in a magneto–optical trap (MOT) even without a specific PA laser under certain condition. In Ref. [21], Caires *et al.* measured the molecular formation rate constant of Rb_2 directly in an MOT. Fioretti *et al.*[14] also mentioned the phenomenon of the formation of Cs_2 molecule in the absence of a PA laser and attributed it to the trapping and the repumping lasers. However, they did not specifically study the properties of Cs_2 molecules produced directly in MOT. Furthermore, the direct ionization process via trapping laser also occurred in experiment of Rydberg atoms,[22,23] during which the trapping laser was used in the first step of excitation process. Thus, the relevant research of the directly ionization process via trapping laser is very important for the fields of ultracold molecules and Rydberg atoms.

In this paper, ultracold Cs_2 molecules in ground electric-state by PA due to the trapping and the repumping beams in MOT are detected by the resonantly enhanced multiphoton ionization technology. We measure the characteristic time of the ground electric-state cesium molecules and investigate the Cs_2^+ molecular ion intensity as a function of the trapping laser intensity and the ionization pulse laser energy. This is useful to enhance the production and trapping of cold cesium molecules using appropriate experimental parameters.

2. Experimental setup

A simplified experimental setup is shown in Fig. 1. An MOT[24] is produced in a stainless steel chamber with a high vacuum environment of about 10^{-7} Pa. Several windows are for optical access and for positioning of a micro-channel plate (MCP). A pair of anti-Helmholtz configuration current-carrying coils provides a 15-Gs/cm (1 Gs=10^{-4} T) magnetic gradient. The trapping beam and the repumping beam are provided by two Littrow external-cavity diode lasers (DL100), respectively. The repumping beam frequency is stabilized to the resonant position of the $6S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ of Cs D_2 line. The trapping beam is tuned about 15 MHz to the red of the $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$ of Cs transition line. Then it is divided into three pairs of mutually orthogonal laser beams each with a power of about 10 mW with opposite $\sigma^{+/-}$ polarizations using $\lambda/4$ waveplates and one of them is overlapped with repumping beam. Two acousto–optic modulators allow a fast shutting of the trapping beam and repumping beam at the same time. We trap about 3.4×10^7 atoms in the Cs cloud with about 0.3-mm diameter and the temperature of Cs atoms sample is about 170 μK.

A pulsed dye laser, pumped by the second harmonic of an Nd:YAG laser (8-ns pulse duration and 10-Hz repetition rate), acts as an ionization laser. In the experiment, the pulsed dye laser is operated at a wavelength of 714 nm. We change the pulse energies within 0–3 mJ and focuse the dye laser beam onto the trapped sample with a waist of the order of 1 mm.

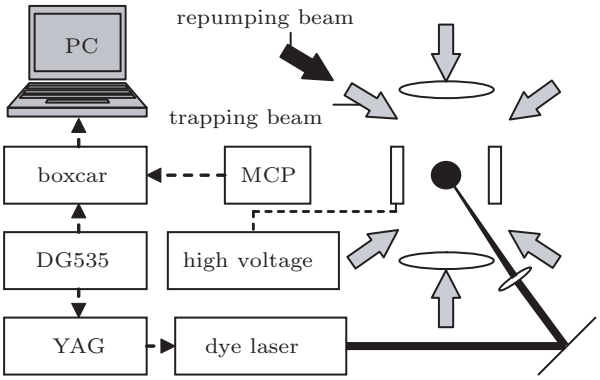


Fig. 1. Experimental setup.

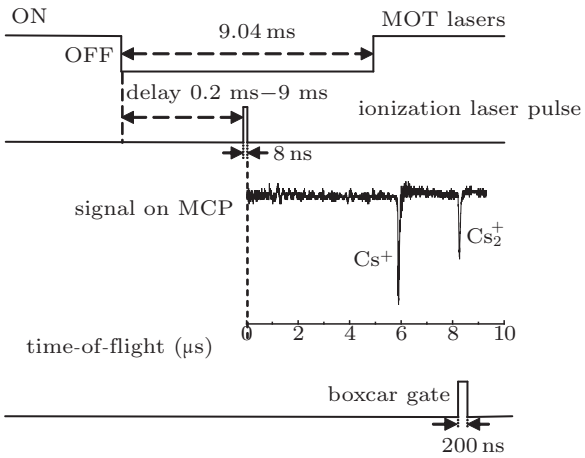


Fig. 2. Time sequence in experiment and the signals of Cs^+ atomic ions and Cs_2^+ molecular ions. This sequence has a repetition of 10 Hz. A typical signal on MCP shows that the Cs^+ and Cs_2^+ signals appear at about 5.9 μs and 8.3 μs after the pulse dye laser, respectively.

The time sequence for the experiment is shown in Fig. 2. An external delay/pulse generator (SRS, DG 535) provides the master clock and controls the switch on/off of MOT lasers, pulsed dye laser and boxcar. A high voltage field (400 V) is applied to the centre of the

MOT by means of a pair of grids spaced 38 mm apart to push atomic ions and molecular ions to MCP, which is located behind one of the grids. Both detected ion signals are processed by a boxcar integrator (SRS-250) and recorded by a computer.

3. Experimental results and discussion

In Fig. 2, we show a typical time-of-flight (TOF) spectra of Cs^+ atomic ions and Cs_2^+ molecular ions. To the best of our knowledge, this is the best signal-to-noise radio of Cs_2^+ ion produced directly in MOT. The position of the Cs^+ appears at $t_{\rm atom} = 5.9$ μs after the pulse dye laser applied and the second well-defined peak appears with a time delay, $t_{\rm mol} = \sqrt{2}t_{\rm atom}$, i.e, at about 8.3 μs, which shows the evidence for the formation of Cs_2 molecules.

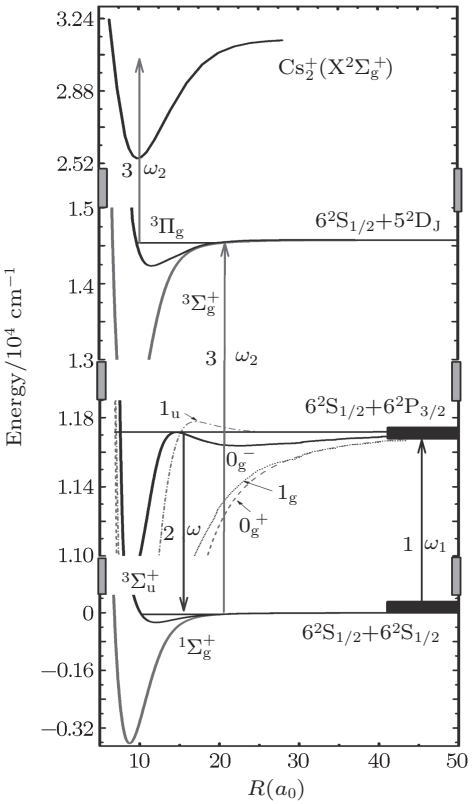


Fig. 3. Formation and detection scheme of ultracold ground electric-state Cs_2 molecules produced directly in MOT.

The formation and the detection scheme of cold molecules produced by PA can be represented by the sequence (see Fig. 3),

$$\mathrm{Cs}(6\mathrm{S}_{1/2}) + \mathrm{Cs}(6\mathrm{S}_{1/2}) + \hbar\omega_1 \xrightarrow{1}$$
$$\mathrm{Cs}_2(6\mathrm{S}_{1/2} + 6\mathrm{P}_{3/2}) \xrightarrow{2} \mathrm{Cs}_2(6\mathrm{S}_{1/2} + 6\mathrm{S}_{1/2}) + \hbar\omega,$$
$$\mathrm{Cs}_2(6\mathrm{S}_{1/2} + 6\mathrm{S}_{1/2}) + 2\hbar\omega_2 \xrightarrow{3} \mathrm{Cs}_2^+(\mathrm{X}^2\Sigma_{\mathrm{g}}^+) + \mathrm{e}^-. \quad (1)$$

In step 1, a pair of ultracold ground state Cs atoms absorbs resonantly a cesium trapping laser photon $\hbar\omega_1$ and photoassociate into Cs_2 molecule in an excited state. In the next step, a fraction of the excited state Cs_2 molecules will spontaneously decay into the ground electric-state vibrational levels of $X^1\Sigma^+$, or the lowest triplet state $a_3\Sigma_u^+$ after emitting a photon $\hbar\omega$. Step 3 is the detection process which is a two-photon, single-colour transition by the pulsed laser (ω_2), promoting the ground electric-state Cs_2 molecules, via an vibration levels of $^3\Pi_g$ state correlated with the $6^2S_{1/2}$+5^2D_J dissociation limit, to a bound levels of the Cs_2^+ potential.

Figure 4 shows the Cs_2^+ molecular ion intensity as a function of trapping laser intensity. The Cs_2^+ intensity presents a linear dependence on trapping beam intensity, which is consistent with the PA process during which one photon from the trapping beam is required [see step 1 in Fig. 3]. Bohn and Julienne[25] provided a model of trap loss induced by PA based on scattering theory, stimulated rate of atoms, Γ, is dependent on PA laser intensity I as expressed below

$$\Gamma = \frac{4\pi^2}{c} I \left|\langle\phi/f_0\rangle\right|^2 d^2, \quad (2)$$

where ϕ and f_0 are the bound excited state wave function and the initial free state wave function in PA process, respectively and $|\langle\phi/f_0\rangle|^2$ represents the free-bound Frank–Condon factor for this transition; d is the molecular dipole matrix element, which is regarded as a constant here. From formula (2) it follows that Γ increases with PA laser intensity I. The number $N_{\rm mol}$ of cold molecules is directly dependent on Γ. The number $N_{\rm ions}$ of Cs_2^+ ions generated in the experiment is proportional to the number $N_{\rm mol}$ of cold molecules, $N_{\rm ions} = \eta N_{\rm mol}$, where η includes the efficiency of the ionization process and the efficiency of the detection process as well. The efficiency of the ionization process, including both excitation to the intermediate state and the ionization itself, is under the influence of FC factor. A rough estimate of the ionization process efficiency is a few percent, less than 10%. The efficiency of the detection process contains the microchannel plate efficiency, which is about 30%, and the ion recollection rate, about 90%. Based on the theoretic analysis, the Cs_2^+ molecular ion intensity is proportional to PA laser intensity I. The relationship between Cs_2^+ molecular ion intensity and trapping beam intensity in Fig. 4 is in accordance with

the theory.

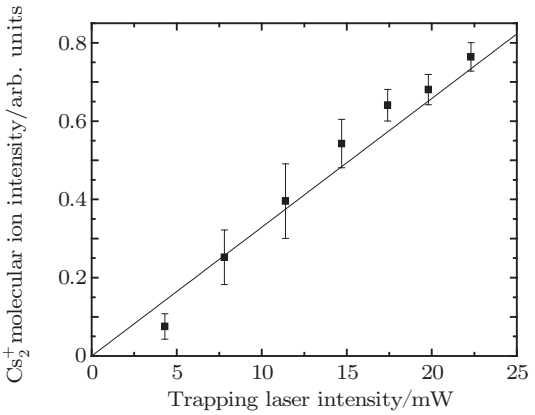


Fig. 4. Cs_2^+ molecular ion intensity as a function of the trapping laser intensity. The solid line denotes a theoretic curve.

By delaying the ionizing laser pulse time starting from the turn-off of the MOT lasers, we observe that the Cs_2^+ molecular ion intensity decays with a characteristic time on the order of 5 ms (see Fig. 5), which is much larger than the radiative lifetime of the photoassociated excited molecules ($\sim$ 20 ns). This result indicates that these ions are produced not by photoassociated excited molecules, but from stable ground electric-state molecules and provides the proof of cold Cs_2 molecules formed in the ground electric-state. In fact, the resulting cold molecular sample is in a ground electric state that has different vibrational states. The observed molecular ion signal in the absence of the PA laser is typically 100 ions/pulse in our experiment. This value is obtained from the characteristic time and amplitude of

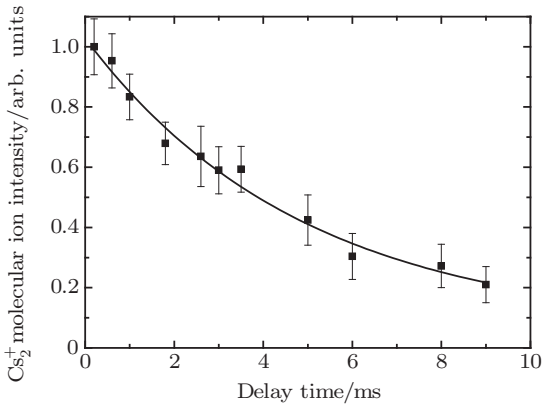


Fig. 5. Cs_2^+ molecular ion intensity as a function of delay time between the ionizing laser pulse and the turn-off of the MOT laser. The solid line denotes the exponential decay fitting with a characteristic time on the order of 5 ms.

a single molecular ion detected on the MCP. Considering that the repetition frequency of the pulsed laser is 10 Hz and the gross efficiency, including the efficiencies of the ionization process and the detection process, is about 2.5%, the molecular production rate in MOT is about 4×10^4 s^{-1}.

As mentioned in Ref. [26], the Cs_2^+ molecular ion intensity is linear with respect to the ionization pulse laser energy and there should appear a saturation effect that follows the increase of pulse laser energy. However, the authors did not observe the saturation phenomenon. In our experiment, where each MOT laser acts as a PA laser, this saturation effect is observed in Fig. 6. We explain it as the fact that because the number of molecules produced by MOT laser is limited, much smaller than that by a specific PA laser, this saturation effect is more easily to take place. In our experiment, errors arise mainly from the fluctuation of pulsed dye laser. The error bars in our experimental results correspond to standard deviations in mean value, averaged over 5 repeated experimental measurements. Errors in a range of 1 mJ–3 mJ are larger than those in the range of 0 mJ–1 mJ in Fig. 6, which is due to the fact that the cross section of the pulsed dye laser and the corresponding pulse fluctuation become large in the case of high energy of pulse dye laser.

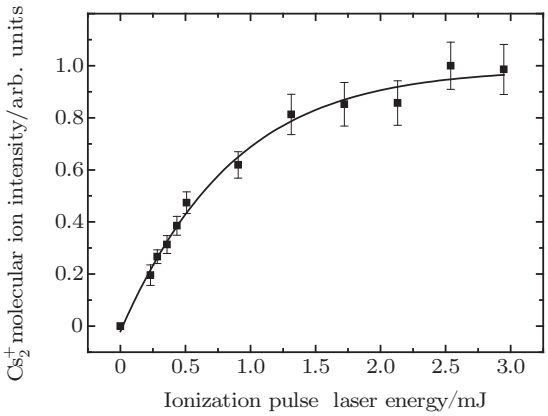


Fig. 6. Cs_2^+ molecular ion intensity as a function of ionization pulse laser energy. The fitting curve shows that the ion intensity is linear when the ionization pulse laser energy is low and that a saturation effect appears when the pulse laser energy is large.

4. Conclusion

We produce and detect ultracold ground electric-state cesium molecules in an MOT where the trapping and repumpping beams act as PA lasers. The production rate of the ultracold Cs_2 molecule is estimated to be up to 4×10^4 s^{-1}. The characteristic time of ultracold ground electric-state cesium molecules is measured to be on the order of 5 ms. The Cs_2^+ molecular ion intensity is linearly dependent on trapping laser

intensity which is consistent with theory. Our experimental results show that the Cs_2^+ intensity is linear when the ionization pulse laser energy is in a range of 0 mJ–1 mJ and a saturation effect appears when the pulse laser energy is over 1 mJ. We hope that our experimental work is conducible to further theoretical and experimental developments in the field of producing ultracold molecules by photoassociation.

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