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Observation of cold state-selected cesium molecules formed by stimulated Raman photoassociation

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We demonstrate the formation of cold ground-state Cs_2 molecules (presumably in a single well-defined rovibrational level) by stimulated Raman photoassociation (PA) in a Cs vapor cell magneto-optical trap. Autler-Townes measurements allow us to precisely define the experimental conditions. The efficiency of the process is comparable to that of the formation of cold molecules obtained after spontaneous deexcitation of photoassociated molecules. Stimulated Raman PA should provide samples where the molecules are cold not only translationally, but are also in a selected rovibrational level.

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To form a sample of cold ground-state molecules, all in a chosen well-defined rovibrational level will really constitute an exciting tool for molecular physics, and also for cold collisions or molecule optics and interferometry. To be able to explicitly detect these molecules is a key for the development of further experiments. Several methods to obtain cold molecules have recently been demonstrated. The first one was the formation of cold molecules via photoassociation (PA) of cold atoms [1]. Another technique consists in magnetic trapping of ground-state CaH molecules [2], cryogenically cooled by ^{3}He buffer gas. Recently, electrostatic loading of ND_3 molecules has been obtained after slowing of an adiabatically cooled beam of polar molecules by time-varying inhomogeneous electric fields [3]. The formation of cold molecules through PA is up to now the only method to reach the microkelvin range. In a PA process, two free colliding atoms resonantly absorb one photon to produce an electronically excited molecule in a well-defined rovibrational level. By exciting the 1_u and 0_g^- states correlated to the dissociation limit $(6s+6p_{3/2})$, PA of cold Cs atoms leads, after spontaneous deexcitation of the photoassociated molecules, to the formation of translationally cold Cs_2 molecules, in either the ground state or in the lowest triplet state, respectively [1]. Temperatures in the range of 20–200 μK (identical to the initial atomic temperatures) and formation rates up to 0.2 molecules per second and per atom have been measured [4]. The molecule formation efficiency depends on both the PA efficiency and the branching ratio (generally weak) between bound-bound (formation of cold molecules) and bound-free (dissociation) transitions for the photoassociated excited molecules. Cesium presents a very efficient scheme because of the particular double-well shape of the photoassociated long-range states, 0_g^- and $1_u(6s+6p_{3/2})$, which provides a Condon point at intermediate distances (15–25 a.u) [1]. The formation of translationally cold molecules via PA has also been observed for rubidium [5], and potassium [6]. Production of cold K_2 molecules has also been achieved via two-step PA through highly excited molecules [7].

The molecules formed via PA followed by spontaneous emission are indeed translationally cold, but they are not internally cold. They are in several rovibrational levels, determined by the Franck-Condon factors. A way around this problem is to use stimulated Raman PA, where the emission on a given bound-bound transition is stimulated (see scheme of the relevant levels in Fig. 1), and which forms state-selected cold molecules [8,9]. Stimulated Raman PA has already been observed in a Rb condensate, where molecules are presumably formed, as indicated by the lineshape of the process only observed through atom losses [10]. Our experiment differs from this performed with a condensate by two main aspects. First, the atomic density in a condensate is three orders of magnitude larger than that of the Cs magneto-optical trap (MOT) sample used, which does not make our experiment a trivial extension of the results of reference [10]. The possibility to obtain an efficient formation of cold molecules via stimulated Raman PA with a low-density atomic sample is not at all obvious. Moreover, compared to the condensate experiment, the achievement of the stimulated Raman PA in a standard MOT opens a simple way for a large range of experiments with cold molecules, when the quantum coherence of the medium is not required. Second, we detect explicitly the molecules by photoionization and time-of-flight mass spectrometry. We report in this Rapid Commu-

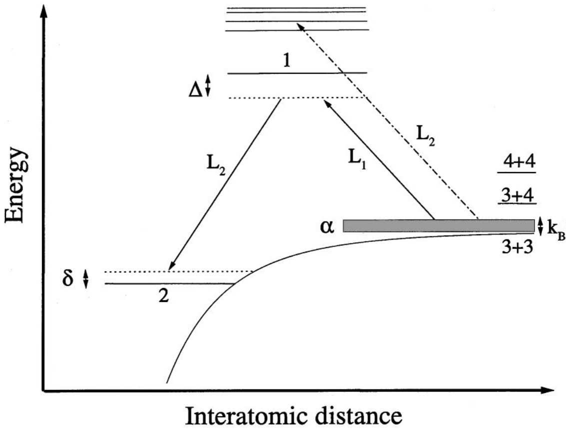

FIG. 1. Relevant energy levels of the stimulated Raman PA transition, from a continuum of states α of the two colliding atoms to a final bound level (2) of the molecule. Δ and δ correspond to the detunings of lasers L_1 and L_2, compared to the PA transition $\alpha\rightarrow 1$ and to the stimulated Raman PA transition $\alpha\rightarrow 2$. The laser L_2 can also produce one-photon PA.

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*Email address: pierre.pillet@lac.u-psud.fr nication the detection of state-selected cold molecules formed in their ground-state via stimulated Raman PA, and we present clear estimates of the amount of so-formed cold molecules.

The principle of the experiment consists, for a pair of colliding cold Cs atoms prepared in the hyperfine level $f=3$, in having a Raman two-photon transition to form directly a ground-state $X^1\Sigma_g^+$ molecule in a well-defined final rovibrational level v,J. The intermediate level, labeled 1 in Fig. 1, is a hyperfine-rotational component of the vibrational level $v=0$ or 1 of the 1_u state, which will be specified later [11]. The first laser, labeled L_1, is detuned by a value Δ either on the red side or on the blue side of the resonance of the PA transition

$$2\mathrm{Cs}(6s,f=3)+h\nu_1\rightarrow\mathrm{Cs}_2^*(1),\tag{1}$$

so that no PA due to the laser L_1 can be observed. We scan the frequency of the laser L_2 to make resonant ($\delta=0$) the Raman transition towards the final ground-state level, labeled 2. One has

$$2\mathrm{Cs}(6s,f=3)+h\nu_1\rightarrow \mathrm{Cs}_2(X^1\Sigma_g^+(6s,f'+6s,f'');v,J)+h\nu_2.\tag{2}$$

The details of the experimental setup are described in several references (see, for instance, [12]). The cold atoms are provided by the use of a Cs vapor-loaded magneto-optical trap. The number of atoms in the trap is 4×10^7 with a peak density of the order of 10^{11} atoms/cm^3, at a temperature $T=130\pm10$ μK. We use the following temporal sequence to prepare the atoms in the hyperfine level $f=3$. At time $t=0$, we switch off the repumping laser for 2.5 ms, to transfer all the atoms from the $6s_{1/2}$, $f=4$ hyperfine level to $f=3$. To force an instantaneous transfer, a few milliseconds before $t=0$ we switch on a pulsed depumping laser (intensity ~1 mW cm^{-2}) tuned to the atomic transition $6s_{1/2};f=4\rightarrow6p_{3/2};f=3$. The presence of the trapping and depumping laser beams does not affect the PA experiment because of their low intensities (<10 mW cm^{-2}). Stimulated Raman PA is achieved by applying on the atomic cloud, continuously the laser beam L_2, and at $t=0$, a laser beam L_1 for a duration $\tau=2.5$ ms. This time is chosen to avoid a too significant decrease of the atomic density. The laser L_1 beam is provided by a Ti:sapphire laser pumped by an argon-ion laser. The available intensity in the MOT zone is up to 450 W cm^{-2}. The laser L_2 beam is provided by a distributed Bragg reflector (DBR) diode laser with a maximum available intensity of 50 W cm^{-2}. The spectral widths of the lasers L_1 and L_2 are, respectively, 1 MHz and 3 MHz. By these spectral widths no further stabilization of the two lasers against each other is necessary to fulfill sufficiently well the two-photon resonance for the Raman process. For observing the formation of cold molecules, we used photoionization of the translationally cold Cs_2 molecules into Cs_2^+ ions, which are detected with a pair of microchannel plates. Ionization is obtained using a pulsed dye laser (applied at $t=2.5$ ms, duration: 7 ns energy: 1 mJ) pumped by the second harmonic of a Nd:YAG laser, running at 10-Hz repetition rate. The dye laser is tuned to the wavelength $\lambda_2\sim716$ nm. The photoionization process for stable molecules is a REMPI process (resonance enhanced multiphoton ionization), via the vibrational levels of an electronic molecular state correlated to the $6s_{1/2}+5d_{3/2,5/2}$ dissociation limits. The REMPI spectrum obtained by scanning the photoionization laser frequency depends on the final rovibrational levels of the ground-state molecules formed. This property has not been analyzed up to now, but makes the molecule detection potentially state selective.

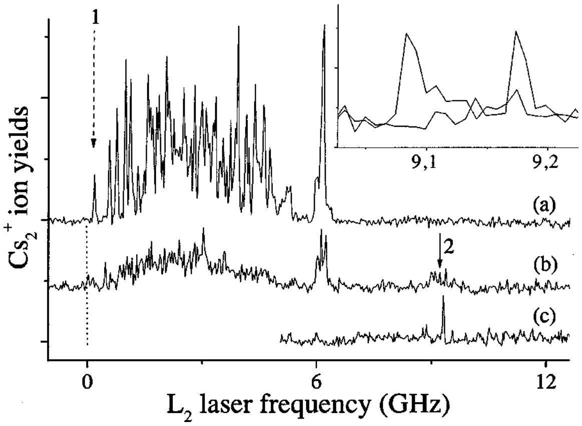

FIG. 2. (a) PA spectrum of the 1_u $v=1$ level, by scanning laser L_2. (b) same as (a) but in presence of the laser L_1. (c) stimulated Raman PA resonance. The inset analyzes the Raman character of resonance (c), for detuning of laser L_1 equal to $+/-40$ MHz (respectively at right and at left).

Figure 2(a) shows the PA Cs_2^+-ion spectrum obtained by applying only the DBR laser (L_2) with an intensity of 30 W cm^{-2}, and by scanning its frequency around the resonance corresponding to one-photon excitation (process shown by the dot-dashed arrow in Fig. 1) of the vibrational level $v=1$ of the $1_u(6s+6p_{3/2})$ state. One has

$$2Cs(6s;f=3)+hv_2\rightarrow Cs_2^*(1_u(6s+6p_{3/2});v=1).\tag{3}$$

The spectrum shows a well resolved relatively complex structure due to the hyperfine interaction and the rotation, which has been analyzed in detail in reference [11]. The maximum of detected ions is here of the order of a few hundred per shot. The dashed arrow indicates in this spectrum a very well resolved and isolated line. This resonance corresponds to the excitation of the hyperfine level, labeled 1, with a total angular momentum $F=7$, and with a wave function close to

$$|(6s+6p_{3/2})1_u\Omega,I_t=7,\Omega_I=7;F=7,M_F\rangle.\tag{4}$$

I_t is the total nuclear spin, and Ω_I and Ω are the projections of I_t and of the total angular momentum of the molecule without nuclear spin, $J_t=1$, on the molecular axis, respectively. Spectrum (b) of Fig. 2 has been obtained in the same conditions as previously, but by applying at the same time the Ti:sapphire laser (L_1). The frequency of laser L_1 (see the dotted line of Fig. 2) is now red-detuned by -20 MHz compared to the resonance 1. The Cs_2^+-ion signal due to the so-tuned laser L_1 alone, is 1.5% of the ion signal with this laser L_1 alone tuned to resonance 1, which means an added background of less than 10 ions per shot. This number is comparable to the Cs_2^+-ion background due to the presence of cold molecules in the MOT even without applying any PA laser [1,5,13]. The presence of the so-tuned laser L_1 perturbs the intensities of the lines in the 1_u spectrum, when scanning the laser L_2 frequency. This perturbation is due to further excitation of the 1_u molecules towards highly excited states, which do not lead to detected molecules. Another characteristic of spectrum (b) compared to (a) is the appearance of new broad structures as shown by the full-line arrow. Reducing the intensity of the laser L_2 down to only $2\ \mathrm{W\,cm^{-2}}$ produces no more direct PA signal due to L_2, but maintains the existence of the extra structures that appear now as very narrow (<10 MHz) resonances [see spectrum (c) of Fig. 2], corresponding to the formation of cold Cs_2 molecules in a single well-defined level of the ground state through stimulated Raman PA. The Raman character of the resonance has been tested by red- then blue-detuning by quantities $\Delta \simeq \pm 40$ MHz laser L_1 from resonance 1. The inset in Fig. 2 shows the shift of the resonance by a quantity $\simeq 80$ MHz, showing that the position of the line is fixed by the frequency difference of the two lasers.

This approach is a reasonably efficient way to make state selected cold molecules, but it is clear that finding the accessible rovibrational levels of the ground state is not an easy matter. With this thought in mind and to unambiguously identify the ground-state rovibrational levels, we have developed a simple way to locate the ground-state levels, we now outline. We have performed two-photon PA spectroscopy of the high vibrational levels of the ground ${}^1\Sigma_g^+$, state, as developed in references [14,15]. We tuned laser L_1 to the PA transition. By scanning the frequency of laser L_2 we observe, each time we have a molecular resonance $Cs_2^*(1) \to Cs_2(2')$ (corresponding to the different possible transitions of Fig. 1), a decrease of the Cs_2^+-ion signal. This PA frustration is due to the Autler-Townes effect induced by laser L_2 (see Fig. 3). We have observed 18 different resonances corresponding to the last levels of the vibrational progressions converging to the $3+3$, $3+4$, and $4+4$ dissociation limits $6s,f'+6s,f''$. The final level 2 corresponding to the spectrum of Fig. 2(c) is located 9.13 ± 0.04 GHz below the $3+3$ limit. The precise analysis of these spectroscopic data is in progress and is not the object of this paper. Starting from the $F=7$ level of 1_u, the total angular momenta reached are $F_t = 6$, 7, and 8. At large interatomic distances, we can assume $J=l$, where l is the angular momentum of the collision. The symmetry of the intermediate level 1 implies that only even partial waves have to be taken into account: here mainly the *s*, *d*, and *g*-waves. In the inset of Fig. 3, we have slowly scanned a given line (indicated by the arrow), showing as expected the rotational progression $J=0$, 2, and 4, which can be observed for most of the lines.

The theoretical approach of stimulated Raman PA is a generalization of the perturbative approach of photoassociation by cw lasers [16]. It predicts that all the formed molecules are in a single rovibrational level of the ground state. It has been demonstrated that the perturbative approach agrees with the measured rates for photoassociation and for cold molecule formation [4]. The rate for stimulated Raman PA [spectrum (c) of Fig. 2] is given by

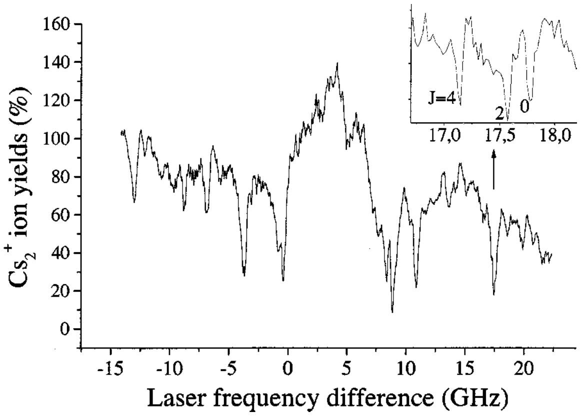

FIG. 3. Two-photon PA spectroscopy analysis of the levels below the $6s+6s$ dissociation limit. The resonances, versus the frequency difference of both lasers L_1 and L_2, appear as a decrease of the Cs_2^+-ion signal (100%) due to laser L_1 on resonance with level 1. More than 100% of signal corresponds to an additional contribution to one-photon PA due to laser L_2. Zero detuning for the laser frequency corresponds to the $f=3+f=3$ dissociation limit. The level 2 corresponding to stimulated Raman PA of Fig. 2(c) is located at 9.13 GHz.

$$R_{RamanPA}^{(1+2)}(\delta) \simeq R_{PA}^{(1)}(\delta) \left| \frac{K_2}{\Delta} \right|^2, \tag{5}$$

where $R_{PA}^{(1)}(\delta)$ is the PA rate to form level 1 due to laser L_1 ($450\ \mathrm{W\,cm^{-2}}$) for a detuning $\Delta' = \delta$. To experimentally determine the Rabi frequency, $2K_2$, for the transition $2 \to 1$, we perform measurements of Autler-Townes splittings, as follows. Laser L_2 is tuned close to the molecular transition $Cs_2(2) \to Cs_2^*(1)$ (see Fig. 1). Laser L_1 is scanned such as to produce the PA process [Eq. (1)]. Figure 4 demonstrates the Autler-Townes effect due to laser L_2. In absence of laser L_2, we observe a narrow resonance peak (a). By applying laser L_2 tuned to the resonance $Cs_2(2) \to Cs_2^*(1)$ with an intensity of $30\ \mathrm{W\,cm^{-2}}$, we observe a splitting of the resonance into two imperfectly resolved peaks (b). Cases (c) and (d) correspond to a detuning of laser L_2 equal to ± 30 MHz. From the splitting of the Autler-Townes resonances of the order of 80 MHz, we deduce a Rabi frequency associated to the bound-bound transition of the order of 75 MHz, for a laser L_2 intensity of $30\ \mathrm{W\,cm^{-2}}$. The Rabi frequency is thus $2K_2 \simeq 19$ MHz for the laser intensity of $2\ \mathrm{W\,cm^{-2}}$. In this rough approach, the number of cold molecules is $N_{CM}^{Raman} \simeq R_{RamanPA}^{(1+2)}(\delta=0)\tau$. This number can be compared to the number of cold molecules produced by laser L_2 ($30\ \mathrm{W\,cm^{-2}}$), $N_{CM}^{PA} \simeq \eta R_{PA}^{(2)}(\delta=0)\tau'$, corresponding to the resonance 1 (dashed arrow) of the spectrum (a) of Fig. 2. $\eta \simeq 0.95$ is the branching ratio for the formation of cold molecules through the 1_u state and $\tau' \simeq 10$ ms the characteristic time of the cold molecules in the MOT zone because of their

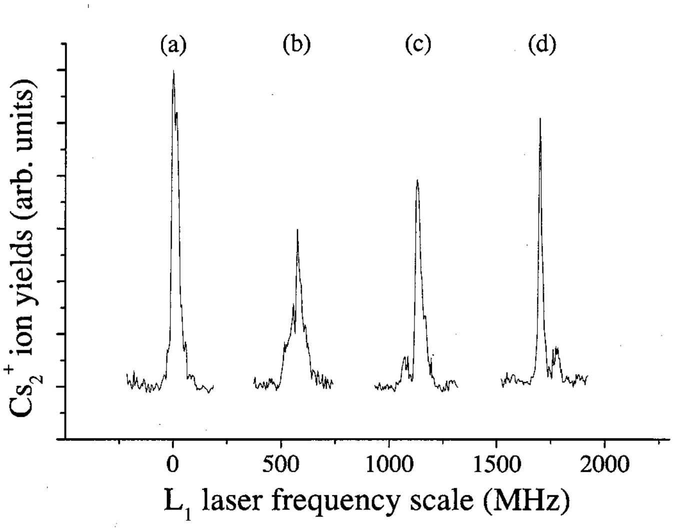

FIG. 4. (a) PA resonance line of the 1_u $v=1$ level (see dashed line of Fig. 2). (b), (c), and (d) are the same resonance with the Autler-Townes effect for different frequencies of laser L_2 (respectively with a detuning equal to zero and $-/+$ 30 MHz) and for an intensity of 30 $W\,cm^{-2}$.

free fall by gravitational force. We expect for the Raman process an ion signal ~ 0.9 smaller, which is about the case here. The stimulated Raman PA signal of Fig. 2(c) corresponds to a number of about fifty to one hundred detected ions, meaning a number of cold molecules of about one thousand, within a factor of three. For $\Delta = \pm 40$ MHz, the Cs_2^+-ion signal is about three times smaller. The width of the Raman resonance is a little narrower than that of the PA resonance, as expected. The resolution is limited by the initial temperature of the atomic sample and the spectral width of the Raman lasers. This peak is assigned to the $J=2$ level. The $J=0$ and 4 levels (separated by ~ 660 MHz) have also been observed, but correspond to weaker resonances, and are hardly visible in the spectrum presented in Fig. 2(c). The theoretical perturbative approach is correct as long as $2K_2 \lesssim \Delta$; above this condition, power broadening is expected. The broad line observed in Fig. 2(b) (see arrow) is the result of the power broadening of the different rotational lines of the reached vibrational level.

To conclude, we have demonstrated the production of translationally-cold ground-state molecules in a well-defined rovibrational level by stimulated Raman PA in a Cs MOT cell. In the weak-field regime, the rate for such a formation of cold molecules can be comparable to the rate of the formation of cold molecules through PA, but now all the molecules are in the same level. This process is really promising. The limit of the efficiency of stimulated Raman PA is the inverse process that should appear when the molecular density become important. Further experiments are to be performed in the near future for the 0_g^- state, as it offers the possibility of preparing cold molecules in the $v=0, J=0$ rovibrational level of the lowest triplet state [17]. Stimulated Raman PA opens the way to prepare molecules that are cold in external and internal degrees of freedom, by using one or several successive Raman processes.

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