Accumulation of Cold Cesium Molecules via Photoassociation in a Mixed Atomic and Molecular Trap

Nicolas Vanhaecke, Wilson de Souza Melo, Bruno Laburthe Tolra, Daniel Comparat, and Pierre Pillet*
Laboratoire Aimé Cotton,† CNRS II, Bâtiment 505, Campus d'Orsay, 91405 Orsay cedex, France

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We have realized a mixed atomic and molecular trap, constituted by a Cs vapor-cell magneto-optical trap and a quadrupolar magnetic Cs_2 trap, using the same magnetic field gradient. We observed the trapping of 2×10^5 molecules, formed and accumulated in the metastable $a\,^3\Sigma_u^+$ state at a temperature of 30 ± 10 μK through a $\simeq 150$ ms photoassociation process. The lifetime of the trapped molecular cloud limited by the Cs background gas pressure is on the order of 1 s.

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The recent developments in the cold-molecule field (for reviews, see [1,2]) require, for many applications, further efforts to obtain a cold, large, and dense sample of cold molecules. Because of the cold molecules' formation rates implied in the photoassociation (PA) schemes, it is necessary to accumulate during a time of the order of 1 s and therefore to keep the molecules in a trap. In this Letter, we report an experimental setup where cold molecules formed via photoassociation are accumulated and trapped.

The formation of cold molecules via photoassociation of cold atoms is, up to now, the only way to prepare a molecular sample in the μK range [3]. The cesium dimer presents very efficient schemes for forming cold molecules either in the ground state or in the lowest triplet state depending on the chosen photoassociated state [3–5]. Temperatures in the range of 20–200 μK [4] (identical to the initial atomic temperature) and formation rates up to one molecule per second and per atom have been measured [6]. The formation of translationally cold molecules through PA has also recently been observed for the potassium dimer using a two-step excitation scheme, and for the rubidium dimer [7–9].

Different molecular traps have already been reported. Trapping of cold molecules present in a Cs vapor-cell magneto-optical trap has been demonstrated using a CO_2 quasielectrostatic trap [10]. As far as we know, no cold molecules formed via PA have been trapped in that experiment. Stimulated Raman PA has been used in a Rb condensate, where the formation of cold molecules is demonstrated through the line shapes of the resonances. These molecules should also be trapped in the magnetic time orbital potential trap used for the condensate [11], but the molecule trapping demonstration was not made. Another technique, quite different, consists of the magnetic trapping of ground state CaH molecules [12] in the tesla range at 400 mK, produced via laser ablation of solid CaH_2 and cryogenically cooled by a ^{3}He buffer gas. More recently, an electrostatic trap and a hexapole torus storage ring loaded with deuterated ammonia molecules, at 350 and 10 mK temperatures, respectively, have been achieved after slowing an adiabatically cooled beam of polar molecules by time-varying inhomogeneous electric fields [13,14].

In our experiment, the molecular cloud produced through photoassociation is in the μK range so that a magnetic gradient in the mT/cm range is sufficient to store it efficiently. The cold atomic cloud, in which PA takes place, is provided by a Cs vapor-cell magneto-optical trap (MOT). In a common MOT, the photoassociated molecules fall down due to the gravity (see, for instance, Fig. 12 in Ref. [4]). But, in the current experiment, the quadrupole magnetic field gradient has been increased to be high enough to compensate earth gravity for molecules, in order to provide both an atomic (magneto-optical) trap and a molecular (magnetic) trap. The cold molecules are formed in the MOT region via PA resonant excitation of two free colliding atoms in a chosen rovibrational level of the $0_g^-(6s + 6p_{3/2})$ state, followed by spontaneous emission towards vibrational levels of the lowest triplet state $a\,^3\Sigma_u^+$ [see Fig. 1(a)]:

$$2\mathrm{Cs}(6s; f = 4) + h\nu_{\mathrm{PA}} \rightarrow \mathrm{Cs}_2^*(0_g^-(6s + 6p_{3/2}); v', J') \quad (1)$$

$$\rightarrow \mathrm{Cs}(a^3\Sigma_u^+(6s + 6s); v) + h\nu_{\mathrm{sp}} \quad (2)$$

In the presence of a magnetic field, the motion of the cold molecules depends on their magnetic moment. Only molecules formed with a magnetic moment opposite to the local magnetic field, in the so-called low field seeking states, can be accumulated and trapped. Because of earth gravity, the other molecules leave the MOT zone and the detection area within 20 ms.

Most of the elements of the experimental setup have been described in previous papers (see, for instance, [15]). The cold atoms in the $6sf = 4$ hyperfine level are provided by a Cs vapor-loaded MOT with residual gas pressure about 10^{-7} Pa. In order to optimize accumulation and trapping, the laser intensities have been reduced and the magnetic field gradient has been increased comparatively to the common MOT operation. The six 8 mm diameter trapping laser beams are tuned 15 MHz to the red of the atomic transition and have a cumulated intensity of 12 mW/cm^2. The magnetic field is of 6 mT/cm, produced by a pair of anti-Helmholtz coils, which is 4 times higher than for the field in a usual MOT. The Gaussian radius of the cold atomic sample is $\simeq 200\ \mu$m, and the number of atoms in the trap is $\simeq 10^7$, leading to a peak density on the order of 10^{17} atoms/m^3. The temperature of the cold atomic sample is estimated to be $\simeq 25\ \mu$K [16,17]. The PA laser is provided by a Ti:sapphire laser (Coherent 899 ring laser) pumped by an argon-ion laser. The laser beam is focused to a $\simeq 300\ \mu$m waist, and the available intensity in the MOT zone is up to 500 W/cm^2. In order to observe the formed translationally cold Cs_2 molecules, they are photoionized into Cs_{2+} ions, which are detected with a pair of microchannel plates through a time-of-flight mass spectrometer. Ionization ($\approx 3\%$ efficiency [6]) is provided by a pulsed dye laser (duration: 7 ns; energy: 1 mJ) pumped by the second harmonic of a Nd-YAG, focused to a 300 μm waist. Most of the data were taken while running the Nd-YAG laser at a 1 Hz repetition rate, limiting to about 1 s the lifetime analysis.

The main results of the experiment are the accumulation and the trapping shown in Fig. 1(b)(1). In this experiment, the atomic and molecular traps run simultaneously. The PA pulse excites, in the MOT region, the $v' = 103$ vibrational level of the $0_g^-(6s + 6p_{3/2})$ state. A few million molecules per second [6] are formed in several vibrational triplet state levels [see Eq. (2)]. While PA is on, the molecules are constantly formed and the 300 ion signal visible at $t = 0$ corresponds to the 10^4 molecules currently present in the detection area. At the end of the PA pulse, nonmagnetically trapped molecules drop out of the 300 μm detection (and MOT) area within $\simeq 20$ ms. The molecules which are clearly detected in the ionization area after this first step are trapped in the shallow quadrupolar magnetic trap. For a molecular temperature similar to the atomic one, the molecular magnetic trap is expected to be roughly a thousand times larger in volume than the MOT, as explained in the later discussion concerning Fig. 3. Therefore, the Cs_{2+} ionic signal left due to the roughly hundreds of molecules present in the ionization area corresponds to an approximately thousand times larger number of molecules actually trapped. The molecular trapping lifetime fitted in Fig. 1(b)(1) is $\simeq 0.15$ s. The influence of the trapping lasers on the molecular trap is shown in Fig. 1(b)(2). The trapping lifetime of the molecular cloud is found to be 4 times longer when the MOT lasers are switched off at the same time as the PA pulse. Then we observe a 0.6 s trapping lifetime as shown in Fig. 1(b)(2).

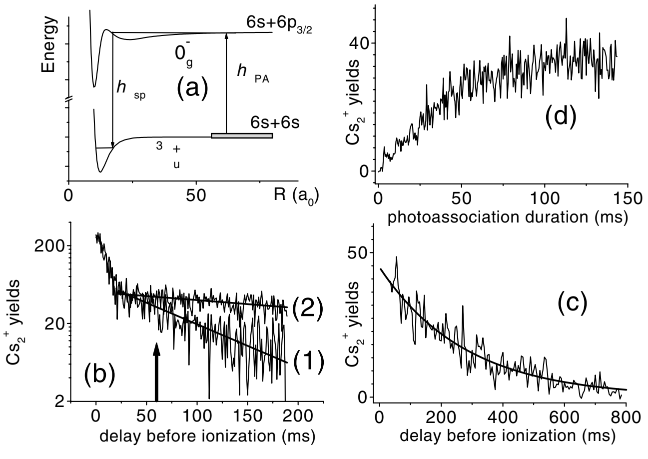

FIG. 1. (a) Schematics of the PA process for the formation of the cold molecules. (b) (1) and (2) In vertical log scale, the evolution of the number of Cs_{2+} ions in the MOT zone after the 150 ms PA process has stopped at $t = 0$: (1) MOT lasers switched on. (2) MOT laser beams switched off. (c) Same as (b2) but with a higher background gas pressure and longer time observation of the trapping, at 0.5 Hz repetition rate and (solid line) exponential decay fitted lifetime. (d) The accumulation process observed 60 ms after the end of the PA process [see arrow in (b)].

The accumulation of cold molecules during the PA is studied by varying the PA pulse length from 0 to 150 ms. The results are shown in Fig. 1(d). The trapped molecules are ionized 60 ms after the end of the PA pulse [see arrow in Fig. 1(b)(1)]. The accumulation of the Cs_2 in the magnetic trap saturates after a PA pulse length of $\simeq 100$ ms. The origin of this effect is linked to the presence of laser light (trapping and PA lasers) during the PA process. Such light can excite and depolarize the trapped molecules, limit;ing the accumulation process. In order to minimize that effect, trapping and PA laser intensities have been adjusted to quite low values, respectively, 12 mW/cm^2 and 35 W/cm^2 for all the spectra presented in this paper.

With such optimized values, we observed the accumulation and the trapping of cold molecules populated by desexcitation of each rovibrational state [$J' = 0$ to 6 in Eq. (1)] reached through photoassociation [18] when the rotational structure is resolved. Figure 2 shows spectra of rotational progressions for two vibrational levels, obtained by detecting the molecules 60 ms after switching off the PA laser (see arrow in Fig. 1). An important observation is the disappearance of trapping in all the rotational levels for any magnetic field gradient below 2.5 mT/cm. It corresponds approximately to the magnetic field gradient $-m_{Cs}g/\mu_B \approx 2.3$ mT/cm necessary to compensate the gravity g for one Cs atom in the hyperfine Zeeman sublevel $f = 4$, $m_f = 4$, which has a magnetic moment given by the Bohr magneton $\mu_B = q_e\hbar/(2m_e)$. Such a result is explained hereafter by evaluating the Zeeman energy of the trapped molecular states.

The trapped cesium dimers are in the lowest triplet state, which is an interesting molecular state due to its large (second order) spin-orbit effect [19], and its even larger hyperfine structure. The spin-orbit coupling which is large at a very small internuclear distance becomes roughly

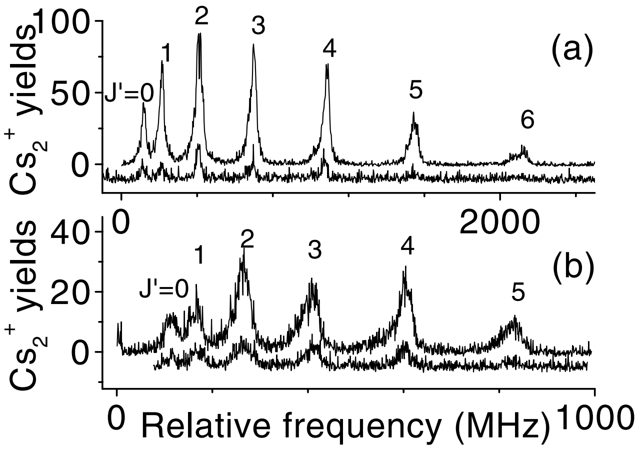

FIG. 2. Trapped molecules for the rotational progressions of $v' = 6$ (a) and $v' = 55$ (b), with a 6 mT/cm magnetic field gradient. For every figure (a) and (b), data of the above are taken at $t = 0$ ms when the PA laser is turned off, and $t = 60$ ms (underneath curve) corresponds to the time given by the arrow in Fig. 1(b).

equal to the hyperfine coupling at the very bottom of the potential curve. The rotation, on the order of $\hbar^2/(2\mu R^2)$, becomes larger than the spin-orbit effect at larger internuclear distances than $\simeq 18a_0$, but remains still smaller than the hyperfine structure. In the current experiment, we photoassociate atoms towards the $0_g^-(v', J')$ state, the desexcitation of which occurs at the classical inner turning point $R_- \simeq (22\text{–}17)a_0$ for the vibrational levels with resolved rotational structure ($v' = 0\text{–}74$) [18,20]. Consequently, for the so-formed cold molecules, the spin $\vec{S}$ is always coupled to the nuclear momentum $\vec{I}$, but not strongly to the rotation of the molecule; this is the Hund's case $b_{\beta S}$ [21,22]. Considering that the Zeeman interaction for two atoms in the s state in a magnetic field $\vec{B}$ is given by

$$H^{\mathrm{Zeeman}} = -2\frac{\mu_B}{\hbar}\vec{S}\cdot\vec{B}, \tag{3}$$

the maximal value of the Zeeman energy is reached when $\vec{S}$ lies along the magnetic field. This maximal value corresponds to molecular states with a magnetic momentum which is twice the Bohr magneton almost independently of the rotational level. Since the molecular mass is twice the atomic one, this description is in reasonable agreement with the experimental results, indicating the trapping of all the rotational levels with the same magnetic field gradient as needed for one atom.

Figure 3 shows the spatial analysis of the trapped molecular cloud at time $t = 50$ ms, done by sweeping the ionization laser horizontally at given vertical positions (compared to the MOT location). The shape of the molecular cloud is a 2 mm horizontal and 6 mm vertical "pear" shape, the peak density of which is located $\simeq 1$ mm below the MOT zone. By integrating the ion signal on the whole molecular gas, and assuming an ionization efficiency near 3% [6], the total number of molecules is evaluated to be 2×10^5. Taking into account a cold-molecule formation rate of the order of 1 molecule per atom and per second, a hundred ms range accumulation process is the only way to explain the experimental number of trapped molecules. Because of the complex hyperfine structure description, the populations of the different rovibrational sub-Zeeman levels in the triplet state are unknown. The temperature of such a distribution is constrained by the maximal value of the molecular magnetic momentum of $2\mu_B$, which gives a maximal temperature of 40 μK. The values of the magnetic momenta of the trapped molecules lie between zero and that maximal value, so that averaging over the Boltzmann distribution of the molecular sample leads to an effective temperature of 30 ± 10 μK.

As mentioned above, the lifetime of the molecular cloud is 0.6 s. It may be limited by collisions with trapped species, by nonadiabatic transfer in a nontrappable state (Majorana transitions), by the radiative lifetime of the molecules, or by collisions with the residual background gas. The molecular trap is so large that the Majorana transitions would limit the lifetime in the range of several hours [23]. Similarly, in the trap, molecules (or atoms) are so diluted (densities $<10^{14}$ m^{-3}) that the collisional processes between the trapped species would limit the lifetime to several minutes (estimated by taking a maximum unitary value for the collisional cross section). These two processes are therefore negligible. Another less obvious effect limiting the lifetime of the molecules inside the trap is the spontaneous emission of the lowest triplet state of the cesium dimer. Indeed, due to the second order spin-orbit effect, the $a\,^3\Sigma_u^+$ state has admixtures from excited states (as $B\,^1\Pi_u$) resulting in a nonzero dipole moment with the ground state $X\,^1\Sigma_g^+$. Spontaneous decay of an

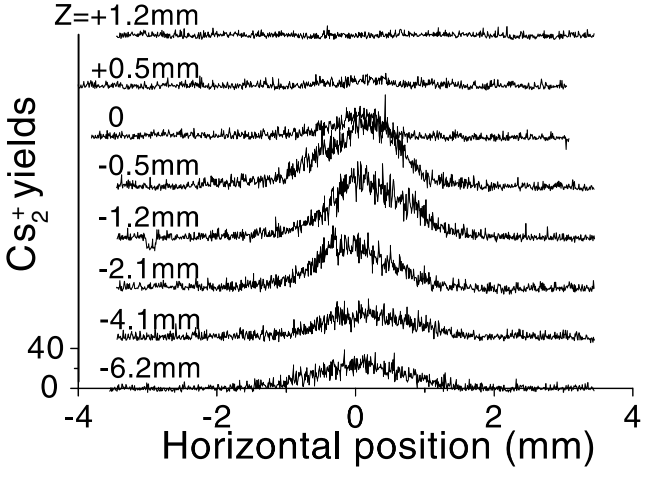

FIG. 3. Molecular cloud analyzed at different positions compared to the MOT position. Z gives the vertical position, and the MOT is located at $Z \simeq 0$. All the vertical scales are the same.

$a^3\Sigma_u^+$ vibrational level towards a $X\,^1\Sigma_g^+$ is therefore possible. In Ref. [24] a value on the order of 100 s is given for rubidium. A scaling argument leads to a lifetime in the range of tens of seconds for cesium, larger than the measured lifetime of the molecular trap. By considering that the residual vacuum corresponds essentially to Cs atoms at room temperature, we estimate a density $n_{\text{res}} \approx 10^{14}$ atoms m^{-3} (which has effectively been measured by absorption) and an average velocity $\bar{v} \approx 200$ m/s. Taking a lifetime $\tau_{\text{res}} = (n_{\text{res}}\sigma\bar{v})^{-1}$ close to the experimental value of 0.6 s leads to a collisional cross section σ ranging 10^{-17} m^2, close to the atom-atom one ($\sigma_{\text{at}} \approx 3.10^{-17}$ m^2 [25]). In order to discriminate between all these possible effects, we have varied the background gas pressure to check if the storage time does depend on it. As an example, Fig. 1(c) shows a trapping lifetime of about 0.4 s, taken with the same experimental conditions as in Fig. 1(b)(2), except for the background gas pressure, which has been increased. A more detailed study has been done and has shown that indeed, in our current setup, the background gas limits the storage time below 1 s. A better vacuum is then needed to be able to observe the radiative lifetime of the triplet state.

To conclude, accumulation and trapping are essential to achieve samples with large numbers of cold molecules. We have demonstrated a convenient tool formed by a mixed atomic and molecular trap. The setup is constituted in a Cs vapor-cell MOT with magnetic field gradient high enough to trap the Cs_2 dimers. The presence of the laser beams limits the accumulation process around 100 ms and therefore the number of trapped cold molecules. The lifetime of trapped cold molecules is actually limited by the background gas below 1 s. We observed 2×10^5 molecules in the lowest triplet state in a 2 mm × 6 mm trap at a temperature to about 30 μK. Further improvements could be investigated, for instance, the use of a denser atomic cloud in order to enhance the PA and cold molecules' formation rate. It would also be interesting to use this mixed trap to store cold molecules in a selected rovibrational level of the lowest triplet state via multiphoton photoassociation schemes [8,26]. This way, we could expect to obtain a large and dense sample of cold molecules in a single rovibrational level, useful for studies of cold collisions, evaporation process, molecule optics, and interferometry or coherent control.

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*Email address: pierre.pillet@lac.u-psud.fr

†Laboratoire Aimé Cotton is associated with Université Paris-Sud.
Electronic address: www.lac.u-psud.fr

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