Atom-Molecule Collisions in an Optically Trapped Gas

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Cold inelastic collisions between confined cesium (Cs) atoms and Cs_2 molecules are investigated inside a CO_2 laser dipole trap. Inelastic atom-molecule collisions can be observed and measured with a rate coefficient of $\sim 2.6 \times 10^{-11}$ cm^3 s^{-1}, mainly independent of the molecular rovibrational state populated. Lifetimes of purely atomic and molecular samples are essentially limited by rest gas collisions. The pure molecular trap lifetime ranges 0.3–1 s, 4 times smaller than the atomic one, as is also observed in a pure magnetic trap. We give an estimation of the inelastic molecule-molecule collision rate to be $\sim 10^{-11}$ cm^3 s^{-1}.

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Cold molecular gases have established a novel field of research in the past few years. After demonstrations of several methods to prepare cold samples of molecules, spectacular advances have been achieved, one of them being the formation of molecular condensates from atomic Fermi gases [1,2]. Increasing interdisciplinary efforts are motivated by a wide range of new phenomena and applications [3]; e.g., cold polar molecules are candidate systems for precision measurements [4] and for quantum information schemes [5].

Understanding and controlling the collisional properties of mixed atomic and molecular gases is crucial to achieve the regime of quantum degenerate molecular gases. Up to now, very few experimental data have been available, and systematic studies of stored atoms, molecules, or mixture of both, have just started [6–8]. As a recent highlight, the formation of ultracold Cs_4 molecules by Feshbach collisions of Cs_2 molecules has been observed [9].

Although a large variety of techniques for the production of cold molecular samples is currently being developed [3], sub-mK temperatures are only reached by photoassociation (PA) out of an initially ultracold atomic sample [10], and by molecule association through magnetic Feshbach resonances [11]. The two techniques are complementary with respect to the nature of the formed molecules: While Feshbach association leads to loosely bound molecules occupying the most weakly bound rovibrational levels, PA allows to populate deeper bound rovibrational states.

Cold molecular samples can be stored using magnetic trapping as demonstrated with Cs_2 [12] and KRb [13] in their triplet state. Trapping molecules in any state is possible in an optical trap as demonstrated by using a CO_2 laser (quasielectrostatic trap, QUEST) with Cs_2 molecules present in a magneto-optical trap (MOT) [14] or formed through a Feshbach resonance [9] and with Rb_2 molecules formed via PA [15].

In this letter, we report the realization of a CO_2 -laser-based QUEST for mixed samples of Cs atoms and Cs_2 molecules. The Cs_2 molecules are formed in the electronic ground state or in the lowest triplet state via PA of cold trapped atoms and are also efficiently trapped inside the QUEST. Lifetime measurements are performed for both atomic and molecular samples. We characterize the inelastic collisions between cold atoms, between cold atoms and cold molecules, and even between cold molecules inside the QUEST. Quantitative data analysis gives us access to the collision rate coefficients.

The basic experimental setup has been previously described (see, e.g., [12]). The main change here is the implementation of a CO_2 laser to realize the QUEST. The cold atoms are provided by a Cs vapor-loaded MOT with a residual gas pressure in the range of 10^{-7}–10^{-8} Pa. The shape of the atomic cloud is approximatively spherical, with a radius of 300 μm. The number of atoms in the magneto-optical trap (MOT) is 10^7, leading to a peak density of 2×10^{10} cm^{-3} by assuming a Gaussian density distribution. A cw CO_2 laser (Synrad Firestar f100) is focused into the MOT zone with a waist of ~ 80 μm at an available power of ~ 110 W.

The temporal sequence for the loading of the QUEST from the MOT is the following. The CO_2 laser is permanently on. First we cool the atoms with an optical molasses phase of 15 ms by switching off the magnetic field gradient and by red-detuning the cooling laser from 2.5 to 27Γ relative to the atomic transition $6s_{1/2}, f = 4 \rightarrow 6p_{3/2}, f' = 5$; here $\Gamma \approx 2\pi \times 5.2$ MHz corresponds to the decay rate of the $6p_{3/2}$ state. The efficiency of the transfer of atoms from the MOT to the QUEST is typically around 3%. The detection of the atoms is performed by a resonant three-photon ionization process via the level 10 s, using a pulsed dye laser ($\lambda = 706.7$ nm) pumped by the second harmonic of a Nd:YAG laser. To prepare the atoms in the hyperfine level $f = 3$ ($f = 4$), we switch off the repumping beam 3 ms before (after) the end of the molasses phase.

Figure 1 shows the temporal evolution of the number of atoms in the QUEST, $N_{Cs}(t)$. For the hyperfine ground state level $f = 3$, we observe an exponential drop with a lifetime of 2.7 ± 0.2 s, limited by the background vacuum pressure. However, for $f = 4$ hyperfine level, inelastic two-body collisions lead to a clearly nonexponential decay at short times. Assuming a Gaussian distribution of the atoms and considering the binary collision parameter

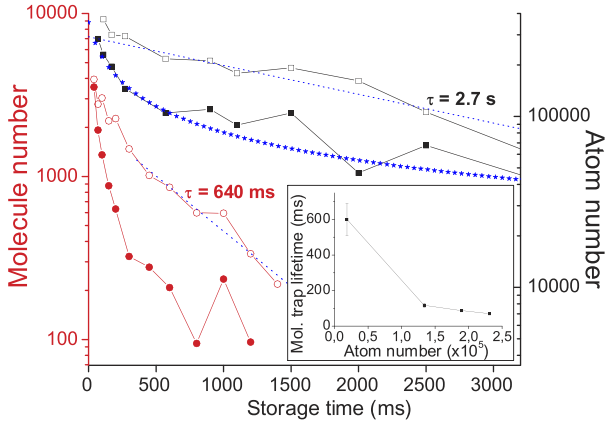

FIG. 1 (color online). Temporal evolution of the atomic and molecular population in the QUEST. The evolution of the number of trapped atoms prepared in the hyperfine states $f = 3$ (open squares line) and $f = 4$ (solid squares line) is plotted. The decay of atoms in $f = 3$ is fitted by an exponentially decreasing function (dot line) with a time constant $\tau = 2.7$ s, whereas, for atoms in $f = 4$, a fit function accounting for two-body collisions (solid stars) is necessary. The solid circles curve shows the evolution of the number of molecules in a mixed atomic and molecular trap. In the inset, we plot the decay constant associated with the molecular trap for different numbers of atoms in $f = 3$ left in the QUEST. We also represent the evolution of a pure molecular sample (open circles line), fitted by an exponential decay function (dotted line) with a time constant $\tau = 640$ ms. In these experiments, molecules are formed via PA of the state $0_g^-(6s + 6p_{3/2})(v = 6, J = 2)$.

$G_{\mathrm{Cs}} = 1.1 \pm 0.3 \times 10^{-11}$ cm^3 s^{-1} [16], we were able to determine an average density $\bar{n}_{\mathrm{Cs}}(0) = 4^{+4.4}_{-2.4} \times 10^{11}$ cm^{-3} after loading the trap. Considering similar atomic QUEST experiments [16], we assume an atomic trap temperature $T \approx 40^{+10}_{-10}$ μK. The number of atoms is then $N_{\mathrm{Cs}}(0) \approx 3.5 \times 10^5$.

The cold trapped atoms are photoassociated in a chosen rovibrational level (v, J), of state $\Omega^{+,-}_{u,g}$, converging towards the electronically excited limit $6s + 6p$

$$2\mathrm{Cs}(6s, f) + h\nu_{\mathrm{PA}} \longrightarrow \mathrm{Cs}_2(\Omega^{+,-}_{u,g}(6s + 6p_{1/2,3/2}); v, J).$$

We have considered different states, 0_g^-, $1_u(6s + 6p_{3/2})$ and $0_u^+(6s + 6p_{1/2})$, leading, for u symmetry (g symmetry), to the formation of cold molecules in a bunch of rovibrational levels (v', J') in the ground state, $X\,^1\Sigma_g^+$ ($v' \sim 132$ after PA of 0_u^+) (in the lowest triplet state, $a\,^3\Sigma_u^+$) [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 $\simeq 300$ μm spot with an available intensity of 300 W cm^{-2}. The maximum number of formed molecules in the QUEST is obtained by applying the PA laser during 30 ms. For longer times, the number of molecules decreases due to the excitation of the molecules by the PA laser. In order to detect the translationally cold Cs_2 molecules, we photoionize them into Cs_2^+ ions (REMPI process), and selectively detect them with a pair of microchannel plates through a time-of-flight mass spectrometer. Molecular photoionization is provided by the same pulsed dye laser as the one used for atomic detection, but using a slightly different wavelength, around 712 nm. We detect an initial number of molecules $N_{\mathrm{Cs}_2}(0) \approx 4000$, corresponding to an average molecular density $\bar{n}_{\mathrm{Cs}_2}(0) \approx 10^{10}$ cm^{-3}.

Figure 2 shows typical PA spectra obtained in a MOT and in a QUEST, corresponding to the $0_g^-(6s + 6p_{3/2}) \times (v = 6, J)$ excitation. For the MOT spectrum the rotational levels $J = 0$ to 3 are shown, and for the QUEST only $J = 2$ level was recorded in this figure. We observe a 42 MHz shift of the resonance between the two spectra because of the dynamical Stark effect due to the CO_2 laser. The expected value of this shift is given by $\Delta\nu = (\alpha_{6s} - \alpha_{6p_{3/2}})P/(\pi h\varepsilon_0 c w_0^2)$, where α_{6s} and $\alpha_{6p_{3/2}}$ are the electrostatic polarizabilities of atoms in $6s$ and $6p_{3/2}$ states [18], P is the power of the laser beam and w_0 its waist. Considering an available power of 110 W, we find $w_0 = 84 \pm 4$ μm, leading to an atomic potential depth of 0.9 mK and a molecular one of 1.5 mK, calculated with the Cs_2 static polarizability given in [19]. The molecular temperature is roughly identical to the atomic one since the PA process does not induce any additional heating [10]. We want to mention here that the analysis of the PA line shape (see fit in Fig. 2) allows us to observe the magnetic sub levels structure of the $J = 2$ state whose degeneracy is lifted by the CO_2 laser electric field [20,21]. This leads to a PA line broadening of ≈ 5 MHz in addition to the natural linewidth of ≈ 10 MHz. The full analysis of the PA line shape is here in agreement with a temperature of ≈ 40 μK.

Figure 1 shows the temporal evolution of the number of atoms and molecules in the trap. The lower curve of this figure (solid circles) shows a lifetime of 110 ms for the molecular cloud in presence of cold atoms, which is quite short compared to the 2.7 s lifetime of the atomic cloud. This molecular trap lifetime does not depend on whether

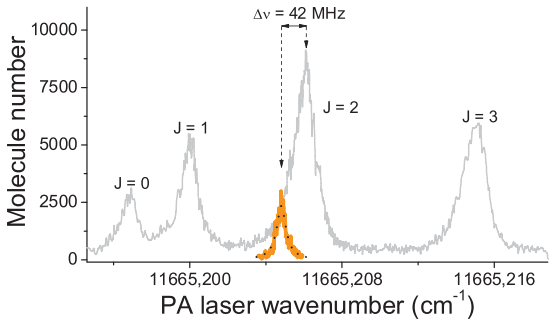

FIG. 2 (color online). PA spectrum of the $0_g^-(6s + 6p_{3/2})$ $(v = 6, J)$ lines in the MOT (gray line) and in the optical trap [dark line (orange)]. A fit of the latter is also represented (dotted line) (see text).

the atoms left in the trap are in $f = 3$ or $f = 4$ hyperfine state. Similar lifetime measurements are obtained after PA of atoms in any of those two hyperfine states. We interpret this lifetime as the result of inelastic collisions between cold atoms and cold molecules. By pushing the atoms outside the QUEST, using a resonant laser beam on the transition $6s_{1/2}, f = 4 \longrightarrow 6p_{3/2}, f' = 5$ (the repumping beam is on again during this phase), we observe an increase of this lifetime up to 640 ms. The inset of Fig. 1 shows the evolution of the molecular sample lifetime versus the number of atoms left in the QUEST. We can see that this lifetime is quite sensitive even to a small number of atoms in the trap. The dynamics of the molecular population for the mixed trap is given by the rate equation

$$\frac{dN_{Cs_2}(t)}{dt} \simeq -[\Gamma_{Cs_2} + K_{Cs_2Cs}\bar{n}_{Cs}(t)]N_{Cs_2}(t) - 2G_{Cs_2}\bar{n}_{Cs_2}(t)N_{Cs_2}(t),$$

where Γ_{Cs_2} is the background loss rate, K_{Cs_2Cs} and G_{Cs_2} are the atom-molecule and molecule-molecule collision rate coefficient, respectively. The molecular population decay is analyzed assuming that the loss of molecules occurred mainly due to collisions with cold atoms, in $f = 3$ hyperfine level. Assuming a constant atomic density, it results an exponential decay of the molecules number. We extract from our data (inset of Fig. 1) the parameter $K_{Cs_2Cs} = 2.6^{+4}_{-1.3} \times 10^{-11}$ cm^3 s^{-1}. This uncertainty on the value of K_{Cs_2Cs} comes from the uncertainty on the atomic density previously determined.

As already mentioned, PA in the potentials 0_g^-, $1_u(6s + 6p_{3/2})$, and $0_u^+(6s + 6p_{1/2})$ was also performed, as shown in Fig. 3(a). We have also considered a particular scheme for the formation of molecules [see Fig. 3(b)], where atoms are photoassociated into one level ($v = 103$) of the outer well of the $0_g^-(6s + 6p_{3/2})$ potential, coupled by tunneling effect with those of the inner one and predicted to form deeper bound molecules ($v' = 7$–9 [22] whereas after PA via $v = 6$ we obtain cold molecules in $v' = 33$–47 [23]) in the triplet ground state [24]. Cold atom-molecule collision rate and molecule collision rate with background gas does not seem to depend on the photoassociated state.

All observed lifetimes of a pure molecular trap are about 4 times smaller than those observed for the atomic cloud ($f = 3$) in the same conditions of vacuum background. To explain this difference, we have first tested the role of the CO_2 laser intensity on both trap lifetimes. We have considered two different waists (84 and 100 μm) for the QUEST leading to two different laser intensities. We have not observed any difference for the atomic and molecular trap lifetime for both waists. It does not seem that the CO_2 laser induces significant transitions between rovibrational levels which are electric-dipole allowed due to the contribution of the second-order spin-orbit interaction [25]. It seems that the explanation for this factor 4 comes

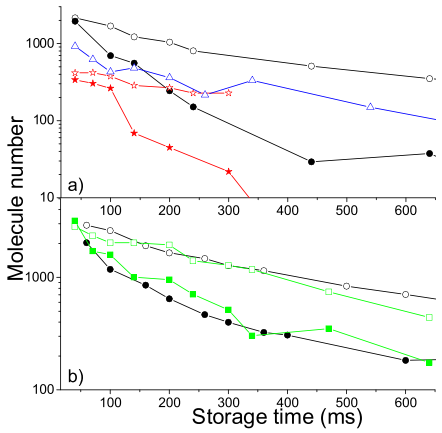

FIG. 3 (color online). (a) Trap lifetime of a molecular sample with atoms (solid symbols) and without (open symbols), after PA of the $0_g^-(6s + 6p_{3/2})$ (circles line), $1_u(6s + 6p_{3/2})$ (triangles line), and $0_u^+(6s + 6p_{1/2})$ (stars line) states. (b) Evolution of the number of trapped molecules with atoms (solid symbols) and without (open symbols) after PA of the $v = 6$ (circle symbols) and $v = 103$ (square symbols) level of the $0_g^-(6s + 6p_{3/2})$ state. Except for 1_u state, molecules are formed via PA of atoms in hyperfine level 6 s $f = 4$ ($f = 3$ in the case of 1_u).

from the fact that the cross section for collisions between hot atom and cold molecules is larger by the same factor compared to the hot atom-cold atom cross section. This result is not so surprising if we consider that the formed and detected molecules are in a bunch of rovibrational levels close in energy to the dissociation limit $6s + 6s$. These molecules have large maximum elongations leading to large impact parameters. To confirm this assertion, we have measured the atomic and molecular trap lifetimes in the case of a pure magnetic confinement with a setup similar to the one of [25]. Here the atomic and molecular densities are 2 orders of magnitude lower, and the inelastic collisions between cold species can be ignored. We have seen that both lifetimes are limited by the background gas collisions and are mostly identical to those observed in the QUEST, with the same factor 4 between atomic and molecular case.

The role of molecule-molecule collisions is not dominant in our experiment compared to the one of background gas collisions. Figure 4 gives the evolution of a pure molecular sample with a reduced background gas pressure. The molecules are formed via PA of cold atoms prepared here in $f = 3$ hyperfine level, which provides a higher rate of cold molecules formation. A simple exponential decay function does not fit very well the data. Nevertheless, with a two-body fit function, the data are better adjusted and we estimate $G_{Cs_2} = 1.0(5) \times 10^{-11}$cm^3 s^{-1}, in agreement with results given in reference [9].

To conclude, we have reported the realization and the characterization of an atomic and molecular QUEST which constitutes a quite exciting tool to trap homonuclear molecules. A systematic collisional study inside the QUEST

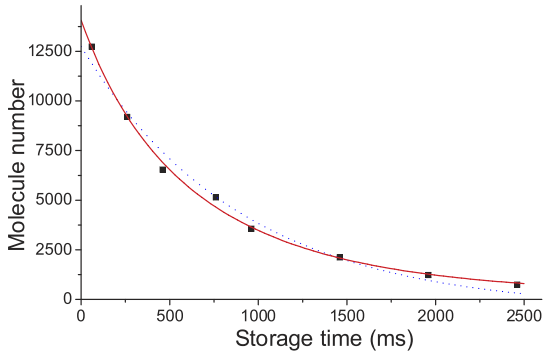

FIG. 4 (color online). Trap lifetime of a pure molecular sample. The evolution of the number of molecules, formed via PA of the $0_g^-(6s + 6p_{3/2})$ $(v = 6, J = 2)$ state, is plotted (solid squares) and fitted with a function accounting for two-body collisions (solid line). A fit with an exponential decay function is also represented (dotted line). The fit yields a lifetime of 900 ms and a two-body rate coefficient of $G_{Cs_2} = 1.0(5) \times 10^{-11}$ cm^3 s^{-1}.

allowed us to determine the atom-molecule and molecule-molecule collision rates. Both rates are close to the unitarity limit ($\approx 1.5 \times 10^{-11}$ cm^3 s^{-1} for atom-molecule collisions and $\approx 8 \times 10^{-12}$ cm^3 s^{-1} for molecule-molecule collisions) and are not compatible with the formation of a stable Bose-Einstein condensate of molecules formed initially from bosonic atoms. The theoretical Ref. [26] predicts an atom-molecule collisional rate quite sensitive to the vibrational level of the formed molecule. In our experiment, we have not seen any evidence of such dependence, but further experiments with selected rovibrational levels are necessary to definitely conclude. Concerning the lifetime of a pure molecular trap, molecule collisions with background gas is the limiting process; the collision cross section being about a factor 4 higher than the one for the atoms. To go further, it can be interesting to increase the trap lifetime by decreasing the vacuum background pressure, which should permit us to measure the lifetime of the metastable molecular triplet state $a^3\Sigma_u^+$, to study more precisely molecule-molecule collisions and to observe molecular transitions induced by the CO_2 laser.

Up to now, the molecules are preferentially formed in high vibrational levels. An interesting challenge is to prepare the molecules in low vibrational levels of the singlet state which may exhibit different behavior against collisional processes. Two-photon PA [27,28] toward highly excited states can offer good configuration. Another challenge is to prepare the molecules in a well defined rovibrational level using Raman PA [27,28], chirped pulse PA [29] or microwave PA. At term a dense and cold molecular sample could be prepared in the lowest rovibrational level $(v = 0, J = 0)$ of the ground state where Bose-Einstein condensation of such a gas would not be impeded by inelastic collisions.

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Note added.—We recently became aware that similar work has been carried out simultaneously in the group of M. Weidemüller at the Physikalisches Institut in Freiburg, Germany [30].

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