Observation of cold Rb_2 molecules trapped in an optical dipole trap using a laser-pulse-train technique

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In this work, we have developed and characterized a laser-pulse-train technique to observe cold Rb_2 molecules trapped in an optical dipole trap. The molecules are produced in a magneto-optical trap, and then loaded into a crossed optical dipole trap. The time evolution of the molecular population is obtained by applying a laser pulse train, which photoionizes the ground-state molecules through intermediate molecular bands. Our results show that this technique allows us to obtain a faster data acquisition rate of the time evolution of the molecule population than other techniques.

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There has been a great interest in the field of ultracold molecules in the last decade, mainly because cold molecular samples are expected to play a major role in different research fields, such as ultracold chemistry, precision metrology, Bose-Einstein condensation, and quantum information [1]. However, the formation of molecules at temperatures below 1 mK can only be performed either by a photoassociation process or by Feshbach resonance or a combination of both processes [1]. After their formation, such molecules can be trapped in an optical dipole trap, since the trapping mechanism does not depend on molecular levels. Depending on the temperature, the molecular sample can be either in a thermal regime [2–6] or quantum regime [7–9]. Furthermore, they can be either in a single vibrational state [7–9] or in a mixture of states [2–6]. In both cases, the time evolution of the molecule population allows one to obtain information on atom-molecule and molecule-molecule cold collisions. Therefore, the detection of such molecules is a key factor in all experiments, and it has been done either through the absorption imaging technique [7–10] or photoionization [2–6,11].

The photoionization technique consists of photoionizing the cold molecules using a pulsed laser beam and observing the charged particle by mass spectrometry. This is a very sensitive and selective technique, allowing the detection of a small number of molecules. This technique has allowed a great development in the field of cold molecules. So far it has been applied using a single laser pulse for each molecular sample, and therefore it requires the preparation of a new molecular sample for each laser pulse [2–6,11]. In order to obtain reliable data, time-consuming averaging of a larger number of measurements over several different molecular samples is required. As a result of such a slow data acquisition rate, the experimental time evolution of the molecules only yields a few data points, limiting the detection rate of molecule-molecule cold collisions. This technique has been applied in this way because it is believed that the photoionization efficiency itself was nearly 100%, although it has never been measured using cold molecules. The development of a faster method of data collection is therefore a timely contribution.

In recent years, our group has been investigating the formation of cold Rb_2 molecules in a magneto-optical trap (MOT) by photoassociation. We have measured the temperature dependence of the Rb_2 molecule formation rate [12], as well as its atomic density and light intensity dependence [13]. In this paper, we have loaded the Rb_2 molecules, formed in an MOT, into a crossed optical dipole trap. In order to observe the molecules, we have developed and characterized a laser-pulse-train technique that enables us to acquire, in real time, the evolution of the molecule population. In addition, we have also measured the ionization efficiency.

Our MOT operates in a stainless chamber with a background pressure below 10^{-9} torr, and it is loaded from a Zeeman-slowed atomic beam. The chamber design is very similar to the one in Ref. [14]. In our setup, we cool and trap either ^{85}Rb or ^{87}Rb atoms. We start from a standard MOT, which traps about 10^8 atoms at an atomic density of 2×10^{10} cm^{-3}. For ^{85}Rb, the trapping laser beam was tuned to the red of the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ atomic transition and the repumping laser beam was tuned to the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ atomic transition. In contrast, for ^{87}Rb, the trapping laser beam was tuned to the red of the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ line and the repumping laser beam was tuned to the $5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F' = 2)$ atomic transition. At the MOT conditions, independently of the chosen isotope, the trapping laser intensity per arm was $I_T = 8$ mW/cm^2 and detuning was $\Delta_T \simeq -3.4\Gamma$, where $\Gamma = 2\pi \times 5.9$ MHz; the repumping laser intensity per arm was $I_R \simeq 1.6$ mW/cm^2 and with a detuning $\Delta_R = 0$. The crossed optical dipole trap is provided by a broadband fiber laser at 1071 nm (IPG Photonics model YLR-40-1070, full width at half maximum of 1.5 nm), which is focused into the MOT volume with a waist of 56 μm at an available power of 25 W at 1071 nm. The experiment runs according to the following time sequence: (i) a MOT loading phase, whose duration is about 4–8 s, consisting of both trapping and repumping laser beams at the initial MOT conditions (the optical dipole trap remains off); (ii) an optical dipole trap loading phase, whose duration is 50 ms. In this phase, the dipole trap beam is turned on and the intensity and frequency of the trapping and repumping lasers are varied to loading conditions ($I_T = 1.6$ mW/cm^2, $\Delta_T \simeq -6\Gamma$, $I_R \simeq 60\,\mu$W/cm^2, and $\Delta_R = 0$). The Rb_2 molecules, formed by photoassociation due to the MOT lasers during this phase [12,13], can be loaded into the optical dipole trap, as in Ref. [3]; (iii) In sequence,

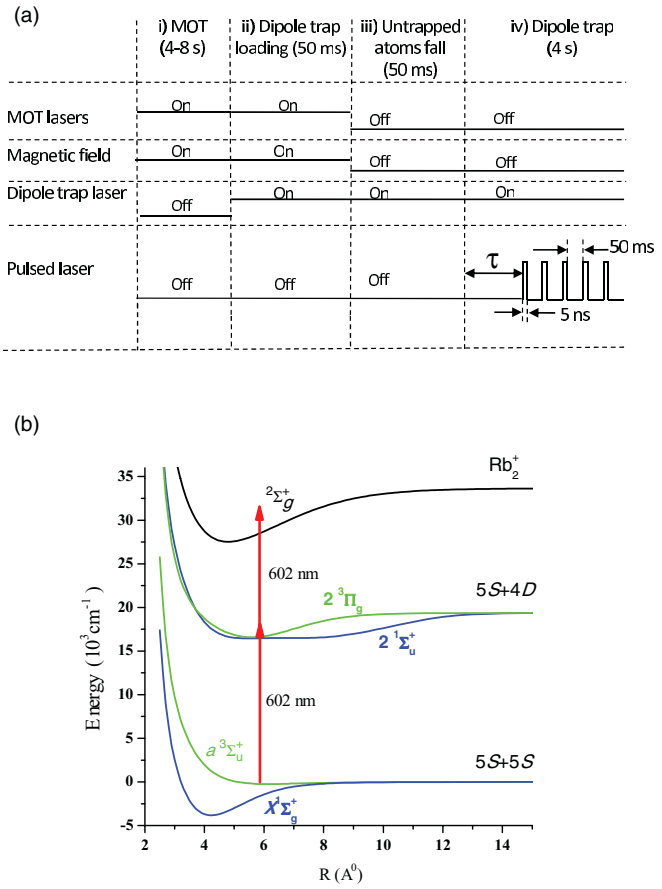

FIG. 1. (Color online) (a) Experimental time sequence. (b) Potential curves of Rb_2 molecule involved in the two-photon ionization process.

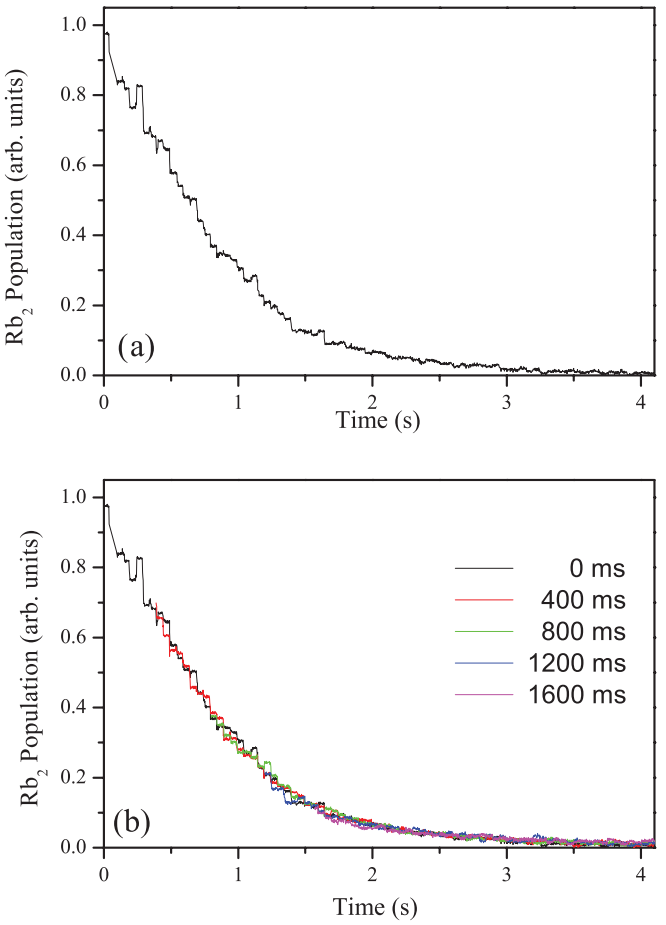

FIG. 2. (Color) (a) Rb_2 population as a function of time, $N_{Rb_2}(t)$, for $\tau = 0$ s. (b) Rb_2 population as a function of time for different holding times into a dipole trap ($\tau = 0$, 400, 800, 1200, 1600 ms). All the curves are normalized to a single common parameter, its initial value $[N_{Rb_2}(t = 0)]$ for $\tau = 0$ s.

the magnetic field gradient, the trapping and repumping laser beams are turned off, and the untrapped atoms are allowed to fall under gravity for 50 ms, leaving behind a pure atomic and molecular sample in the dipole trap. In the typical condition, we have about 3×10^6 trapped atoms at a density of 3×10^{12} cm^{-3}; (iv) Finally, after a holding time (τ) in the dipole trap, a laser pulse train is applied to the sample to photoionize the trapped Rb_2 molecules. Ions are detected by a channeltron electron multiplier. The whole time sequence is shown in Fig. 1(a). The ionization probe laser beam is provided by a pulsed dye laser (Jaguar-Continuum) pumped by the second harmonic of a Nd:YAG laser (5-ns pulse duration, running at a 20-Hz repetition rate), operating at the wavelength of 602 nm. The pulsed dye laser was focused to a Gaussian width of 250 ± 50 μm at the position of the dipole trap. It is important to mention that we are not able to distinguish between the molecules formed in ground singlet and/or triplet states. However, we believe that they should be in the triplet state as suggested in Ref. [15]. The reason for this assumption is that the Franck-Condon factor is larger to connect the 0_g^- state to the triplet than to the singlet ground state. In any case, the photoionization process involves a two-photon transition through the rubidium diffuse $a^3\Sigma_u^+ \rightarrow 2^3\Pi_g$ ($X^1\Sigma_g^+ \rightarrow 2^1\Sigma_u^+$) molecular band for the triplet (singlet) molecules [Fig. 1(b)], well known from early studies in the context of light-induced fluorescence experiments (LIF) [16].

A boxcar gate is used to integrate the ion signal, which is then collected by a digital oscilloscope, and transferred to a computer. The whole data acquisition requires about 80 laser pulses.

To characterize the laser-pulse-train technique, we have performed a few tests using a mixed sample of ^{85}Rb atoms and molecules trapped into an optical dipole trap. In Fig. 2(a), we show the Rb_2 population as a function of time, $N_{Rb_2}(t)$, obtained for at 1-mJ energy per pulse for $\tau = 0$ s. This curve is an average of 10 different curves, and the data were normalized to their initial value $[N_{Rb_2}(t = 0)]$ along the y axis. If the photoionization efficiency is too high, a laser pulse train could deplete the Rb_2 population, leading to a decay curve that includes photoionization losses as well. In order to investigate the importance of such an effect on the molecule population, we have varied the holding time (τ) in the dipole trap before turning on the laser pulse train. If the photoionization losses are important, we should observe different time evolution curves of the Rb_2 population for different τ. In Fig. 2(b), we show the Rb_2 population as a function of time (t). Each curve starts at time $t = \tau$ (0, 400, 800, 1200, and 1600 ms) and is normalized to a single common parameter, its initial value $[N_{Rb_2}(t = 0)]$ for $\tau = 0$ s. From this graph, it is clear that each curve superposes

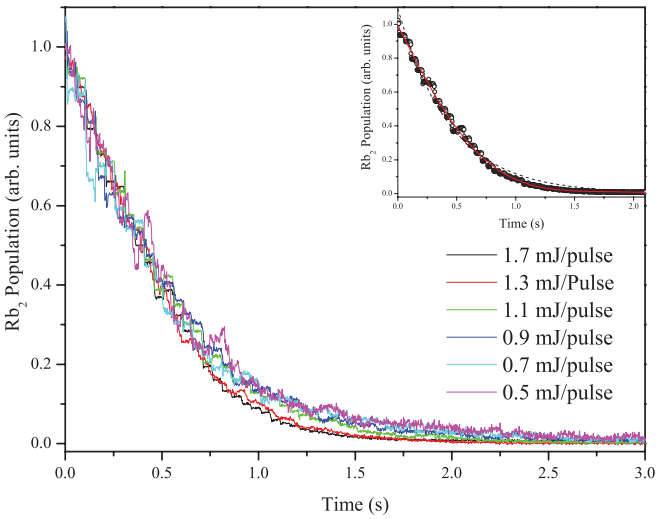

FIG. 3. (Color) Rb_2 population as a function of time for $\tau = 0$ s at different values of energy per pulse of the ionization laser. Each curve was normalized to its $N_{Rb_2}(t = 0)$. The inset shows the Rb_2 population as a function of time for 1.7 mJ/pulse. The dashed line is an exponential fit and the red line is the fitting considering the photoionization losses.

the previous ones, independently of the holding time into the dipole trap. This finding suggests that photoionization losses of the molecular sample are negligible and the molecular sample was not depleted by the ionization probe laser.

In order to investigate the role of the pulsed laser energy on the depletion of the Rb_2 population, we measured $N_{Rb_2}(t)$ as a function of time for $\tau = 0$ s at different values of energy per pulse of the ionization laser (Fig. 3). We varied the energy per pulse in a range where the Rb_2^+ signal is known to be proportional to the square of this parameter [13]. In fact, we verified that $N_{Rb_2}(t = 0)$ as a function of pulsed laser energy presents a quadratic dependency. Each curve in Fig. 3 is an average of 10 different curves, and was normalized to its $N_{Rb_2}(t = 0)$ for comparison. From Fig. 3, we emphasize that (i) the ratio signal to noise improves as the energy per pulse increases, as expected; (ii) the Rb_2 population time evolution is basically the same for 0.5, 0.7, and 0.9 mJ/pulse or, in other words, the ionization losses can be considered negligible for those conditions.

We observed that the $N_{Rb_2}(t)$ curve can be fitted by a single exponential decay only for 0.5, 0.7, and 0.9 mJ/pulse. Such curves, which present no ionization losses $[N^{NL}_{Rb_2}(t)]$, can be described by $N^{NL}_{Rb_2}(t) = e^{-t/\tau_c}$, where τ_c is the decay constant. Since the data is normalized, we do not need to consider $N_{Rb_2}(t = 0)$ in the fitting procedure. This allows us to obtain $\tau_c \sim 500$ ms, for 0.5, 0.7, and 0.9 mJ/pulse. The lifetime is believed to be due to collisions only, either with background gas or atoms and molecules trapped. If we consider the molecular curve that presents ionization losses $[N^{L}_{Rb_2}(t)]$, we will be able to estimate the ionization efficiency per pulse. We will consider that each pulse ionizes a fraction f of the Rb_2 population, so after n pulses the population can be written as $N^{L}_{Rb_2}(t) = e^{-t/\tau_c}(1 - f)^n$. Therefore, besides the collisional losses there are also photoionization losses. If we make the ratio of the two populations at $t = n\Delta t$ (Δt is the time interval between two laser pulses), we will have $N^{L}_{Rb_2}(n\Delta t)/N^{NL}_{Rb_2}(n\Delta t) = (1 - f)^n$. Here, we will consider the data 0.9 mJ/pulse as $N^{NL}_{Rb_2}(t)$ and the one for 1.7 mJ/pulse as $N^{L}_{Rb_2}(t)$. For $t = 1.5$ s ($n = 30$ and $\Delta t = 50$ ms in Fig. 3), the Rb_2 population for 0.9 mJ/pulse is approximately 2.5 times larger than the Rb_2 population for 1.7 mJ/pulse, which gives us $f \sim 0.030 \pm 0.008$. If a fitting is performed for the 1.7 mJ/pulse curve using the $N^{L}_{Rb_2}(t)$ expression, we obtain $f \sim 0.028 \pm 0.007$ (red line in the inset of Fig. 3; the dashed line is an exponential fitting). For comparison, in 50 ms (time interval between laser pulses) the losses due to collisions are about 10% of the whole population, which is larger than the photoionization losses per pulse (3%). In any case, if the collisional losses are smaller than the photoionization losses the population decay will be dominated by the latter. We have also verified that the laser pulse train did not deplete the atomic population by observing its time evolution using the absorption imaging technique. The number of trapped atoms and its time evolution were the same in the presence and absence of the laser pulse train.

The observed molecular ion signal in the dipole trap at 1.7 mJ/pulse is typically 100 ions/pulse for the first pulse. This value is obtained with the characteristic time and amplitude response of our channeltron observed for a single molecular ion. The channeltron's manufacturer specifies its efficiency as about 70%; the ion collection efficiency is estimated to be 95% using ion optics simulation software and it is due to a nickel mesh with 95% transparency in front of the detector. Finally, the ionization process efficiency is 3%. Combining all these data, we find that the initial molecular population is about 5×10^3 molecules in the crossed dipole trap. This population is consistent with the molecule production due to photoassociation during the loading phase [13].

The temperature of the trapped Rb_2 molecules is another important parameter of the sample. To measure it, we have switched off the dipole trap and allowed the molecular sample to expand. After some expansion time, we applied a single ionization pulse. Using this technique, we are able to measure the Rb_2 population which remained in the ionization

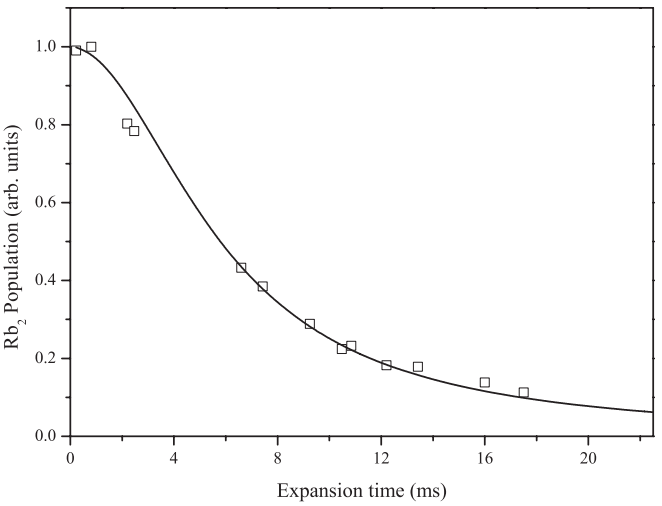

FIG. 4. Rb_2 population as a function of expansion time after being released from the dipole trap. The full line is a theoretical model for 30 ± 10 μK.

region after the expansion, as shown in Fig. 4. The ionization region is determined by the pulsed laser beam waist. A simple numerical model is used to obtain the molecular temperature, which considers a Maxwell-Boltzmann distribution of velocities, a Gaussian spatial molecular distribution, the pulsed laser beam intensity profile, and gravity. Such a model allowed us to deduce a molecular translational temperature for the produced Rb_2 to be 30 ± 10 μK, in accordance with our previous results [12,13]. We should point out that the main source of uncertainty in this measurement is systematic and it is due to the pulsed laser beam waist, which determines the ionization region. The laser beam waist error is an important parameter in the model to evaluate the temperature error bar.

In a final experiment, we investigated the Rb_2 population decay as a function of the atomic density in the dipole trap for both isotopes using 0.9 mJ/pulse. Recent experiments have observed atom-molecule cold collisions in mixed samples trapped in optical dipole traps [2,4]. In our case, we have verified that all the Rb_2 population decay curves, obtained at 0.9 mJ/pulse as a function of the atomic density, can be fitted by a single exponential. This fact allows us to conclude that the molecule-molecule cold collision rate is negligible, and we will not consider it here. Therefore, the time evolution of the molecular density (n_{Rb_2}) in a mixed trap is given by the following rate equation:

$$\frac{dn_{Rb_2}}{dt} = -\Gamma n_{Rb_2}, \quad \Gamma = [\Gamma_b + \beta n_{Rb}(r,t)],$$

where Γ_b is due to background losses involving collisions between hot atoms and cold molecules, β is the atom-molecule cold collision rate coefficient, and $n_{Rb}(r,t)$ is the Rb atomic density. In principle, we would need to know the functional form of $n_{Rb}(r,t)$ to fit properly the decay curves. However, all our data can be fitted with a single exponential with the same lifetime. This suggests that Γ is independent on the atomic density in the mixed dipole trap for both isotopes, as shown in Fig. 5. And, therefore, we do not need to know the decay curve for n_{Rb}. Nevertheless, the atomic sample lifetime

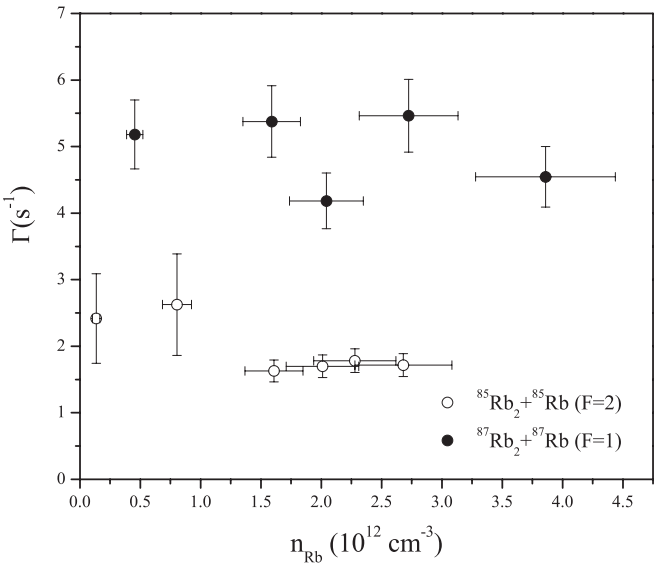

FIG. 5. Γ as a function of the atomic density in the mixed dipole trap for both isotopes.

is larger than the molecular lifetime. Such behavior suggests that collisions between cold atoms and cold molecules are not present in our experiment, and $\Gamma = \Gamma_b$. Nevertheless, there is an isotope difference, because Γ for $^{85}Rb_2$ is about 2.5 times smaller than for $^{87}Rb_2$. Since the molecules are formed by photoassociation, using near resonant light, it is very likely that their population is distributed among high-lying vibrational states of the ground electronic potential, independently of the isotope. Therefore, background collisions should be the same for both samples. A possible explanation for this observation is that the 1071-nm broadband laser is driving bound-to-bound transitions within the molecule to excited electronic states, asymptotically correlated to the $5S + 5P$ state. Subsequently, the molecules decay to lower vibrational levels in the ground electronic state, which are dark states for the ionization laser. In another words, there is an optical pumping process to dark molecular states that appears to induce molecular losses in our molecule dipole trap. This effect could be responsible for the different $\Gamma's$, since vibrational levels of the excited electronic states will occur at different energies for each isotope. If that is indeed the case, Γ should depend on the laser intensity of the optical dipole trap. We have investigated this point and our results show that Γ is independent of such a parameter from 0.1 to 0.6 MW/cm^2. Such independence may be due to the fact that the laser intensity is well above the saturation intensity of the molecular transition. Another interesting observation is that Γ_b for $^{85}Rb_2$ is about four times larger than the background losses for cold ^{85}Rb atoms in the dipole trap, perhaps due to this molecular optical pumping process as well. A more detailed investigation shall be performed in the near future. We should also mention that it is surprising that we did not observe collisions between cold atoms and cold molecules, especially because our atomic density was about 10 times higher than the densities used in Refs. [2,4]. The main difference between the referenced experiments and ours is that our molecules were formed in high-lying vibrational states due to the near resonant light photoassociation. On the other hand, the results obtained in [2,4] were obtained for low-lying vibrational levels ($v = 4$–6 and 32–47) of the triplet electronic ground state. This speculative explanation of the discrepancy remains to be investigated.

In conclusion, we have developed and characterized a laser-pulse-train technique to observe cold Rb_2 molecules in an optical dipole trap. This technique allows us to have a faster data acquisition rate than destructive techniques. Furthermore, it also allows us to obtain reliable photoionization efficiency and molecular temperature. By trapping MOT formed molecules, we observed isotope differences for the background collisional losses. We believe this effect is due to an optical pumping scheme involving excited electronic states of each isotope. Future experiments, with well-defined vibrational levels, should clarify this issue. Furthermore, the pulse-laser-train technique may be important to future experiments involving the trapping of cold molecules, allowing the acquisition of more data at a fast rate.

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