PHYSICAL REVIEW A VOLUME 59, NUMBER 1 JANUARY 1999

Observation of cold ground-state cesium molecules produced in a magneto-optical trap

T. Takekoshi, B. M. Patterson, and R. J. Knize
Laser and Optics Research Center, Department of Physics, U.S. Air Force Academy, Colorado 80840
(Received 18 May 1998)

We have observed translationally cold electronic ground-state cesium dimers produced directly in a magneto-optical trap and also by photoassociation of cold atoms in this trap. These neutral molecules were detected using photoionization and time-of-flight spectroscopy. Initial experiments indicated a cold molecular formation rate of about 600 s^{-1}, with no photoassociation laser present. The measured translational temperature of the neutral cesium dimers was approximately 100 μK. [S1050-2947(99)50401-0]

PACS number(s): 33.80.Ps, 32.80.Pj, 33.80.Eh

Laser cooling is routinely used on the alkali metals, alkali-metal earths, metastable inert gases, and a few transition metal elements, but there has been no demonstration of this technique on molecules. The reason is that additional vibrational and rotational structure makes it much more difficult to realize the high photon scattering rates necessary to reduce the translational kinetic energy of molecules. A special broadband light source would be required to prevent molecules from being optically pumped into ''dark'' states that do not interact with the laser fields [1]. An alternative is to use elastic collisions between room-temperature molecules and a conventionally cooled helium vapor to thermalize the molecules to temperatures on the order of a few hundred mK [2]. Recently, Fioretti and co-workers at Orsay showed that cold molecules can also be formed from laser-cooled atoms [3]. We confirm this in the present paper and demonstrate that translationally cold electronic ground-state molecules can be produced directly from laser-cooled atoms in a magneto-optical trap (MOT). We observe these molecules as a by-product of a normal MOT and also demonstrate their formation using photoassociation.

Our source of laser-cooled atoms was a cesium vapor cell MOT [4]. While we found that a small molecular signal could be observed in standard MOTs under normal trapping conditions, denser MOTs produced a larger molecular signal. Transiently compressing the MOT by ramping up the magnetic-field gradient [5] caused the yield to increase an order of magnitude. Reducing the repumping light intensity (thereby creating a temporal ''dark'' MOT [6]) at the same time increased the signal by another 10%, but is not used in the data presented here. These signals were present several milliseconds after the MOT trapping light was turned off. The signals are due to electronic ground-state ($6S+6S$ asymptotic) molecules because the electronic excited-state ($6S+6P$ asymptotic) dimers formed by photoassociation have a lifetime of 15 ns.

The MOT parameters were as follows: The MOT laser beam diameter was 2 cm, the intensity per beam was 3.5 mW cm^{-2}, and the frequency was detuned by $\delta=-2\Gamma$ from the trapping transition. The background gas pressure was 10^{-9} Torr, which resulted in an atomic trap lifetime of about 4.5 s. The axial magnetic-field gradient was switched from 7 to 25 G cm^{-1} to transiently increase the density. At the high-magnetic-field gradient, the MOT contained about 2.5×10^{6} atoms in a diameter of about 170 μm. Since the compressed MOT was operated in the multiple scattering regime [7], we assumed a uniform atomic density of about 10^{12} cm^{-3}.

The molecules were detected by photoionizing them with light from a pulsed, frequency-doubled neodymium-doped yttrium aluminum garnet laser ($\lambda=532$ nm). The beam entering the vacuum chamber was about 1 mm in diameter at the MOT. The pulses had an energy of about 13 mJ with a pulse width of 3 ns full width at half maximum. At the resulting intensity, the two photon processes for molecular photoionization are saturated. The following ionization events can occur in the MOT [8]:

$$\mathrm{Cs}+2\nu\rightarrow\mathrm{Cs}^{+}+e^{-}, \tag{1}$$

$$\mathrm{Cs}_2+2\nu\rightarrow\mathrm{Cs}^{+}+e^{-}+\mathrm{Cs}, \tag{2}$$

$$\mathrm{Cs}_2+2\nu\rightarrow\mathrm{Cs}_2{}^{+}+e^{-}, \tag{3}$$

$$\mathrm{Cs}_2+2\nu\rightarrow\mathrm{Cs}^{+}+\mathrm{Cs}^{-}. \tag{4}$$

At the wavelength used, the photoionization cross section for Eq. (2) is about the same as that for Eq. (3) and the cross section for the last process is negligible. A timing circuit was used to introduce a variable delay after turning off the MOT beams and before the photoionization laser pulse. The ions were detected directly using a model 4721 Channeltron (Galileo Corporation). The Channeltron input was biased at -2700 V inside the grounded stainless-steel vacuum chamber, allowing time-of-flight spectroscopy to be used. It was mounted inside a stainless-steel tube attached to the main chamber in order to reduce the electric field at the MOT. This resulted in longer times of flight, which allowed the atomic and molecular signals to be resolved.

Figure 1 shows a typical time-of-flight spectrum of ions detected as a function of time after the photoionization laser pulse. For this experiment, a low gradient MOT is loaded for about 250 ms. Then the cold atoms are compressed for 40 ms using a high field gradient to increase the atomic density. In Fig. 1, the photoionization laser pulse occurs at time zero, which is 0.5 ms after the MOT laser beams are turned off. The large signal that occurs at 45.0(2) μs arises primarily from two-photon photoionization of atoms [Eq. (1)]. The smaller signal at 63.8(2) μs arises from ions created from neutral cesium dimers [Eq. (3)]. The ratio of arrival times of

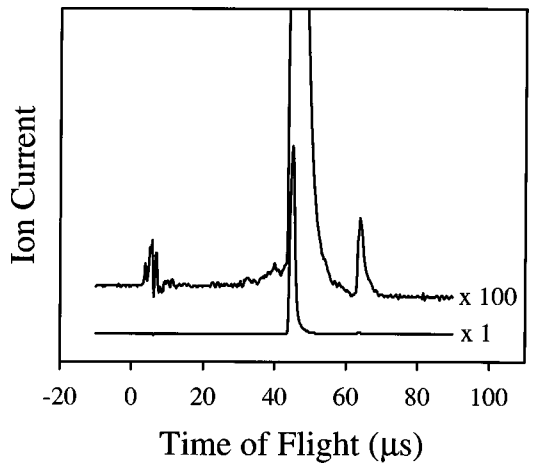

FIG. 1. Time-of-flight spectrum of the detected ions averaged for 200 shots. A large atomic ion signal is observed at 45.0(2) μs after the photoionization pulse. The molecular ion signal observed at 63.8(2) μs corresponds to about three molecules per shot. In these data, the MOT beams were turned off 0.5 ms before the photoionization pulse. The small oscillation at 5 μs is due to an electrical transient.

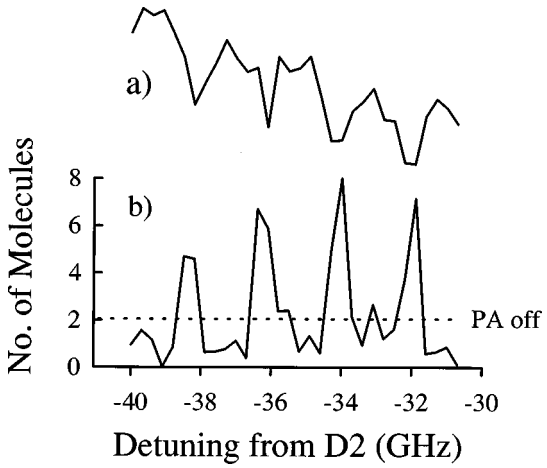

FIG. 2. (a) Precompressed MOT atom fluorescence as a function of the photoassociation laser frequency. Photoassociation is observed as a decrease in fluorescence. (b) Molecular ion signal from the compressed MOT as a function of photoassociation laser frequency. The absolute frequency scale is known only to within ± 3 GHz. The molecular ion signal maxima correlate with the minima of the atom fluorescence. For this curve, the MOT beams were turned off 1.5 ms before photoionization. The dashed line shows the molecule signal obtained in a compressed MOT without the photoassociation laser present.

molecular to atomic ions is 1.418(8), which agrees with the expected ratio of $\sqrt{2}$. The background vapor atomic and molecular ion signals are negligible on the scale shown.

In addition to producing dimers directly in a MOT, we also detected translationally cold neutral molecules produced by photoassociation [9], as has been done very recently by Fioretti and co-workers [3]. Photoassociation experiments usually detect the decrease in the MOT fluorescence or the creation of molecular ions as a function of the photoassociation laser wavelength. Our photoassociation laser was a Spectra Diode Labs model 8630-high power diode laser with the grating and rear high reflector removed. The rear facet of the chip was injected with about 30 mW of light from a single-mode free running Sanyo model 6033 diode laser. The high power chip was operated at a current of 1.2 A, where it acts purely as a single pass amplifier and the output power is nearly constant with wavelength. The output was 150 mW with a linewidth <50 MHz, as measured with a spectrum analyzer, and the beam diameter at the MOT was 0.25 cm. The photoassociation laser was left on throughout the loading, compression, and detection phases. There was a 1.5-ms delay between turning off the MOT laser beams and photoionization. Figure 2 shows the fluorescence of the precompressed MOT as well as the corresponding compressed MOT molecular photoionization signal, as a function of the photoassociation laser frequency detuning from the $D2$ transition. The results confirm that the size of the molecular signal can be enhanced with a photoassociation laser and that the increase in photoassociation directly correlates with an increase in the number of detected translationally cold electronic ground-state neutral molecules.

It is possible that the molecular ion signal we see without photoassociation arises from atomic ions colliding with cold atoms, rather than directly from cold, neutral molecules. This possibility was investigated by removing atoms from the trapping region. The high-power diode laser was tuned to the Cs atomic $D2$ transition, allowing it to be used as an atom removal laser. The MOT was loaded and the trapping beams were turned off. After 2 ms, the atom removal laser was shuttered on for 2 ms to quickly accelerate atoms out of the detection region. Then the photoionization laser was pulsed. Compared to the control situation where there was no atom removal laser, the dimer signal was 0.8 times the control dimer signal, while the atomic signal was 0.06 times the control atomic signal. This means that the molecular ion signal does not arise from ion-atom collisions.

The translational temperature of the ground-state molecules produced through compression alone was measured by varying the delay time between turning off the MOT laser beams and photoionization. During this time, the cold molecular cloud expands according to its temperature and drops due to gravity. Figure 3 shows the detected molecular ion signal as a function of the delay time. The atomic ion signal showed a similar decrease. A simple Monte Carlo model was used to determine the temperature of the molecules and the results are also shown in Fig. 3. This model predicts a cesium dimer translational temperature of 100 ± 100 μK. The temperature of the cesium atoms in the low gradient MOT used for loading is estimated from the MOT population, light intensity, and light detuning [7] to be about 100 μK.

While Fioretti and co-workers have shown that photoassociation of Cs produces mainly triplet ground-state molecules [3], we do not know the fraction of singlet ($^1\Sigma_g^+$) and triplet ($^3\Sigma_u^+$) Cs_2 we produce through simple compression. The exact mechanism is also unknown. Using the molecular translational temperature, the detected ion signal size, the branching ratio, the fact that the molecular photoionization was nearly saturated, and an estimated Channeltron gain, the molecular formation rate without the photoassociation laser was determined to be about 600 s^{-1}. One possible mechanism for formation of cold molecules is three-body recombination of cold atoms. The three-body recombination rate for doubly spin-polarized cesium was calculated [10] to be about

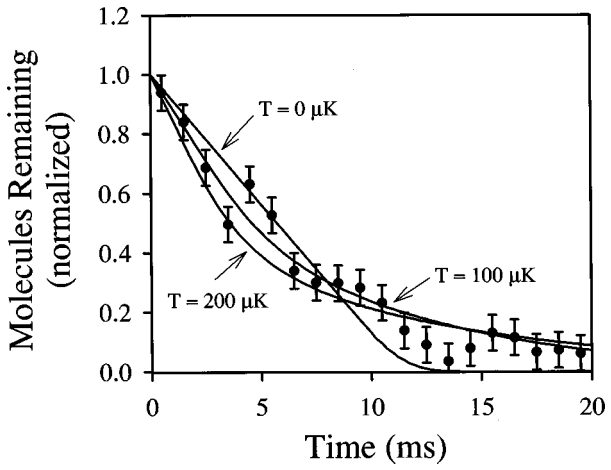

FIG. 3. Relative ion signal arising from cesium dimers as a function of the delay time after the MOT laser beams were turned off. The three solid curves are results from a Monte Carlo simulation. The temporal decay of the signal corresponds to a cesium dimer temperature of about $100 \pm 100\ \mu$K.

5×10^{-29} cm^6 s^{-1}. Using an estimated uniform atomic density of 10^{12} cm^{-3}, this yields a molecular formation rate of about 130 s^{-1}, which is the same order of magnitude as the measured yield. However, photoassociation by the MOT trapping beams is also a possible explanation for our compressed MOT molecule signal. Further measurements and calculations will be required to fully understand the origin of molecules produced directly in a MOT. An obvious first step would be to measure the dependence of the dimerization rate on the atomic density n. The total molecular formation rate scales as n^3 for three-body recombination and as n^2 for photoassociative dimerization. The exact rotational and vibrational energy distributions of the molecules are also unknown. However, both of the mechanisms mentioned here are expected to create molecules with vibrational energies close to the continuum. In the case of photoassociation, this is because the intermediate excited-state molecules will have binding energies close to $\hbar\delta$, where δ is the detuning of the MOT lasers. Spontaneous decays from here to weakly bound vibrational levels are far more probable than to strongly bound ones. In the case of three-body recombination, calculations on Li, Na, and Rb show that the partial cross section for the highest vibrational level always dominates for triplet molecules [11].

We have shown that cold cesium dimers can be produced directly in a MOT and also by photoassociation. The measured temperature of the cold cesium molecules was about 100 μK. The observed molecule production rate was the same order of magnitude as that predicted for three-body recombination of cold spin aligned cesium atoms. These cold, neutral, ground-state molecules could be useful for spectroscopy, frequency standards, and molecular optics.

We acknowledge the support of the U.S. Air Force Academy, the Air Force Office of Scientific Research, the National Research Council, and the Research Corporation.

[1] J. T. Bahns, W. C. Stwalley, and P. L. Gould, J. Chem. Phys. **104**, 9689 (1996).

[2] J. Kim, B. Friedrich, D. P. Katz, D. Patterson, J. D. Weinstein, R. DeCarvalho, and J. M. Doyle, Phys. Rev. Lett. **78**, 3665 (1997).

[3] A. Fioretti *et al.*, Phys. Rev. Lett. **80**, 4402 (1998).

[4] C. Monroe, W. Swann, H. Robinson, and C. Wieman, Phys. Rev. Lett. **65**, 1571 (1990).

[5] W. Petrich, M. H. Anderson, J. R. Ensher, and E. A. Cornell, J. Opt. Soc. Am. B **11**, 1332 (1994).

[6] C. G. Townsend, N. H. Edwards, K. P. Setie, C. J. Cooper, J. Rink, and C. J. Foot, Phys. Rev. A **53**, 1702 (1996).

[7] M. Drewsen *et al.*, Appl. Phys. B: Lasers Opt. **59**, 283 (1994).

[8] J. Morellec, D. Normand, G. Mainfray, and C. Manus, Phys. Rev. Lett. **44**, 1394 (1980); E. H. A. Granneman, M. Klewer, K. J. Nygaard, and M. J. Van der Wiel, J. Phys. B **9**, 865 (1976).

[9] C. C. Tsai *et al.*, Phys. Rev. Lett. **79**, 1245 (1997); J. J. Blange *et al.*, *ibid.* **78**, 3089 (1997); H. Wang *et al.*, Phys. Rev. A **55**, R1569 (1997).

[10] E. Tiesinga, A. J. Moerdijk, B. J. Verhaar, and H. T. C. Stoof, Phys. Rev. A **46**, R1167 (1992).

[11] A. J. Moerdijk, H. M. J. M. Boesten, and B. J. Verhaar, Phys. Rev. A **53**, 916 (1996).