Observation of Optically Trapped Cold Cesium Molecules

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We report the first observation of optically trapped cold neutral molecules. Cesium dimers in the electronic ground state are produced directly in a magneto-optical trap and transferred to a dipole trap formed at the focus of a CO_2 laser beam ($\lambda = 10.6\ \mu$m). These neutral molecules were detected using photoionization and time-of-flight spectroscopy. Initial experiments indicate a cold molecule trap lifetime on the order of half a second. [S0031-9007(98)07817-X]

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There have been many experiments demonstrating the production and trapping of cold atoms. Atoms with temperatures less than 1 mK can be produced using laser cooling on the alkalis, alkaline earths, metastable inert gases, and a few other elements. Once cooled through collisions with photons, these cold atoms can be confined in various optical and magnetic traps and can be used for a variety of applications [1]. In order to create large scattering forces, laser cooling requires a closed two-level transition, or a relatively simple structure that can be closed with the use of a few additional lasers to avoid optical pumping. This requirement has prevented laser cooling of most atoms and of all molecules. There is interest in producing cold molecules for Doppler-free spectroscopy, cold atom-molecule and molecule-molecule collisions, molecular quantum collective effects, alkali cluster studies, frequency standards [2], molecular optics [3], as well as many other applications already realized for atoms. An alternative cooling method is to sympathetically cool molecules by using a cold buffer gas. They can then be confined in a strong magnetic trap. Cold Eu atoms have already been trapped using this method [4], which could be extended to paramagnetic molecules.

An all-optical method for producing cold molecular vapors is difficult because of vibrational and rotational degrees of freedom, which greatly complicate direct laser cooling [5]. However, it was recently demonstrated by Fioretti and co-workers at Orsay [6] that cold electronic ground state Cs dimers can be produced from laser-cooled atoms [7]. We have independently verified this result [8] and report here the trapping of these cold Cs dimers in an optical dipole force trap formed at the focus of a CO_2 laser beam ($\lambda = 10.6\ \mu$m). This demonstration of the ability to trap cold molecules will allow ultracold molecular vapors to be observed for long times without the necessity of large magnetic fields. Many of the interesting experiments already done with laser cooled atoms may be possible with molecules as well.

We produced cold molecules using a vapor cell magneto-optical trap (MOT) [9]. Our initial detection of the cold molecules produced continuously in a MOT was straightforward. The trapping beams were suddenly switched off, and both atoms and molecules were detected using photoionization. A frequency doubled Nd:YAG laser ($\lambda = 532$ nm) producing about 13 mJ in a 3 ns pulse was utilized. This pulse creates atomic and molecular cesium ions by two-photon processes [10],

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

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

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

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

At a wavelength of 532 nm, the cross section for the process given in Eq. (2) is about the same as that for Eq. (3). The cross section for the last process is negligible. The photoionization laser was focused so that the beam diameter was about 0.1 cm at the MOT. 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. We observe an ion signal attributed to molecules because it arrives at a time $\sqrt{2}$ times that of the atom ion signal. The molecules must either be in the triplet ground state (as observed by Fioretti and co-workers [6]) or in the singlet ground state because the delay (a few ms) between turning off the trapping light and photoionizing is much longer than the excited state molecular lifetime (15 ns).

Unlike the Orsay group [6], we do not use a separate photoassociation laser to produce cold electronic ground state molecules. We enhance the small molecular signal seen from a normal MOT by increasing its density. The simplest way to demonstrate this is to suddenly ramp up the MOT's magnetic field gradient. This increased the molecular signal by an order of magnitude, which is expected in light of the fact that the rates of the two most probable mechanisms for dimer formation vary as the atomic density squared (photoassociation by the MOT lasers) and the atomic density cubed (three-body recombination). By varying the delay between switching off the MOT beams and photoionizing, the resulting decay due to falling and expansion is observed. From this, we calculate a molecular translational temperature of about 100 μK. From the temperature and number of molecules detected, we infer a molecular production rate of about 600 s^{-1}, which is the same order of magnitude as the rate calculated using the triplet three-body recombination rate of Tiesinga and co-workers [11].

We have also demonstrated that these molecules can be loaded into a dipole force trap [12]. We use a variant called a quasielectrostatic trap [13]. This type of trap is most easily implemented using a focused $\mathrm{CO_2}$ laser beam ($\lambda = 10.6\ \mu\mathrm{m}$). It is suitable for trapping electronic ground state homonuclear diatomic molecules because vibrational transitions between ground states, as well as dissociative transitions to the continuum, are electric dipole forbidden. Molecules with large permanent electric dipole moments may require alternate optical trapping methods [14]. Photoionization and electronic transitions to excited $\mathrm{Cs_2}$ states require many photons and are expected to be insignificant [15]. The potential energy of the trapped species can be written to good approximation as $U = -\alpha_s E^2/2$, where α_s is the electrostatic polarizability and E is the rms electric field. The relevant polarizabilities for our experiment are $\alpha_s(\mathrm{Cs}) = 59.6 \times 10^{-24}\ \mathrm{cm}^3$ and $\alpha_s(^1\Sigma_g{}^+\mathrm{Cs_2}) = 104 \times 10^{-24}\ \mathrm{cm}^3$ [16] [$\alpha_s(^3\Sigma_u{}^+\mathrm{Cs_2})$ should be comparable].

In our trapping experiment, cold molecules were loaded into the dipole trap by overlapping the waist of the $\mathrm{CO_2}$ beam with a "dark MOT" (Fig. 1) [17]. The MOT beam diameters were 2 cm and the maximum intensity per beam was 3.5 $\mathrm{mW\,cm^{-2}}$. The frequency was detuned by $\delta = -4\Gamma$ from the $6S_{1/2}F = 4$ to $6P_{3/2}F = 5$ cycling transition (852 nm), where Γ is the natural linewidth. Repumping light was provided by a σ^+-σ^- retroreflected 2 cm diameter laser beam tuned to the $6S_{1/2}F = 4$ to $6P_{1/2}F = 4$ transition (895 nm). Because Cs has such a large hyperfine splitting, an additional linearly polarized depumping beam tuned to the $6S_{1/2}F = 4$ to $6P_{3/2}F = 4$ transition (852 nm) was combined with the $\mathrm{CO_2}$ laser beam to further reduce the $F = 4$ ground state population in the dipole trap region, thus increasing the atomic density. The 17 W linearly polarized $\mathrm{CO_2}$ laser beam was focused to a waist radius of about 64 μm and was shuttered with an acousto-optic modulator (AOM). This creates a dipole trap which, including gravity, is about 200 μK deep for Cs atoms and about 350 μK deep for Cs dimers. The dipole trap also has the effect of locally changing the frequencies of the MOT trapping, repumping, and depumping atomic transitions. The atomic $6P$ state is also trapped and because of the neighboring $5D$ state, actually has a larger dc Stark shift than the $6S$ ground state. At the center of the trap, the $D2$ ($6S_{1/2}$ to $6P_{3/2}$) transition was Stark shifted by about -2.9 natural linewidths [18] and the $D1$ ($6S_{1/2}$ to $6P_{1/2}$) transition was Stark shifted by -2.5 natural linewidths [19]. The $\mathrm{CO_2}$ beam was roughly guided onto the MOT by using the depumper as an alignment beam. Finer alignment was achieved by chopping the $\mathrm{CO_2}$ beam with its AOM and maximizing the effect seen on the MOT fluorescence signal due to the Stark shifts, when it was fed into a lock-in amplifier.

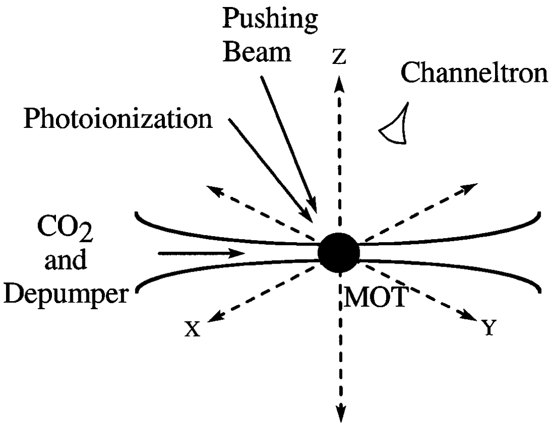

FIG. 1. Schematic of the experimental geometry. The dipole trap beam and depumping beam are combined. Their waists ($\approx$130 μm $1/e^2$ diameter) intersect with the MOT, which is roughly twice as large. The MOT beams travel along the axes shown, with the repumper along the x axis. The "pushing" beam is used to remove atoms from the dipole trap. The trapped atoms and molecules are ionized by the pulsed photoionization beam ($\lambda = 532$ nm) and are then detected by the channeltron.

A typical loading and detection cycle works as follows: About 7×10^6 atoms are loaded into the 200–250 μm diameter MOT with the trapping and repumping beams at maximum intensity. There is no $\mathrm{CO_2}$ or depumping beam present. Then the trapping and repumping beam intensities are reduced at the same time the $\mathrm{CO_2}$ and depumping beams are turned on. This loading period lasts for about 100 ms, after which all of the atoms are pumped into the $F = 3$ ground state by extinguishing the repumping laser for 0.5 ms. Then the MOT trapping laser, the depumping laser, and the magnetic field are all turned off. Some of the atoms and molecules remain in the dipole trap as the untrapped atoms and molecules fall away. After a variable time delay, the atoms and molecules are released. They are then detected by photoionization. Figure 2*a* shows the time-of-flight ion spectrum observed 110 ms after the MOT is turned off. The large signal that occurs at 44.4(2) μs arises primarily from Cs^+ created from two-photon photoionization of atoms [Eq. (1)]. The smaller signal at 62.6(2) μs arises from $\mathrm{Cs_2}^+$ created from neutral cesium dimers [Eq. (3)]. The ratio of arrival times of molecular to atomic ions is 1.410(8), which agrees with the expected ratio of $\sqrt{2}$. Figure 2*c* shows that without the $\mathrm{CO_2}$ laser, both the atomic and molecular signals are negligible.

In order to show that the dimer signal was not a result of continuous molecule formation from three-body recombination of atoms in the dipole trap, the number of atoms was reduced by using a pushing laser beam. This linearly polarized, 1 cm diameter, 1 $\mathrm{mW\,cm^{-2}}$ beam was the same

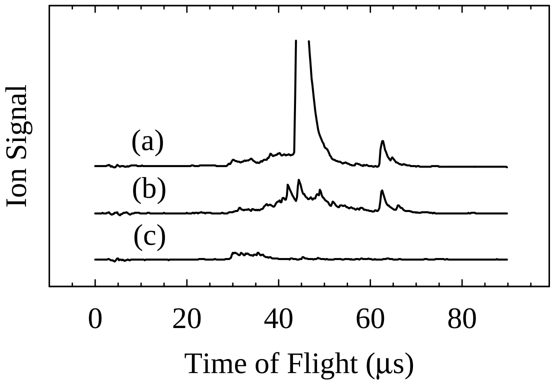

FIG. 2. Time-of-flight spectra of atoms and molecules in the dipole trap (100 shot averages). (*a*) Time-of-flight spectrum 110 ms after the MOT is switched off. The photoionization pulse occurs at 0 μs. A large atomic signal and a small molecular signal are observed at the appropriate times. (*b*) Spectrum observed at 110 ms, when a pushing beam is used to remove the atoms and leave the same number of trapped molecules. The small scattered signals near 44 μs are partly due to atomic ions created by the process in Eq. (2). (*c*) Spectrum observed at 110 ms when the CO_2 laser is not turned on. The residual signal is due to photoionization of Cs on the windows, and Cs from the background gas.

frequency as the MOT trapping light. It was shuttered on (simultaneously with the repumper beam) 50 ms after the MOT was turned off and shuttered off 10 ms later. Figure 2*b* shows that the number of atoms remaining was reduced by at least a factor of 50, while the number of dimers remained about the same. Because of this, we can rule out three-body recombination of trapped atoms as a source for the molecular signal. Using the molecular ion signal size, the photoionization branching ratio, the fact that the molecular photoionization was nearly saturated [10], and an estimate for the channeltron gain at high mass (266 amu), we calculate that the molecular peak in Fig. 2*b* corresponds to something on the order of 6 molecules. The actual number of molecules trapped is probably about a factor of 4 higher, because the photoionization beam illuminates only the central $\frac{1}{4}$ of the trapping volume.

The decay of the molecules from the trap as a function of time is illustrated in Fig. 3. The data show that the molecules remain in the trap for times on the order of 0.5 s. Since this is the same order of magnitude as the atomic trap lifetimes we have observed previously, we assume this is due mainly to collisions with thermal background gas. Further data will clarify the molecular loss mechanism.

In this experiment, the fraction of singlet and triplet state molecules is unknown but could be measured using state selective two-photon photoionization of the ground state molecules. The rotational and vibrational state distributions are also unknown. However, both possible dimerization mechanisms are expected to primarily populate the highest vibrational states [20]. In future experiments, we will measure the density dependence of the molecular formation rate to determine whether photoassociation or three-body recombination is the dominant mechanism for molecule formation. Also, in order to do future experiments on cold molecular vapors, it is important to greatly increase the number of molecules trapped. This can be addressed by use of an additional photoassociation laser in the MOT, as well as by using denser atom traps such as those used to produce Bose-Einstein condensation. It may even be possible to use the recent proposal by Julienne and co-workers [21] to produce molecules directly from a Bose-Einstein condensate. Possible future applications using the optical trapping technique to produce cold molecular vapors are clearly numerous.

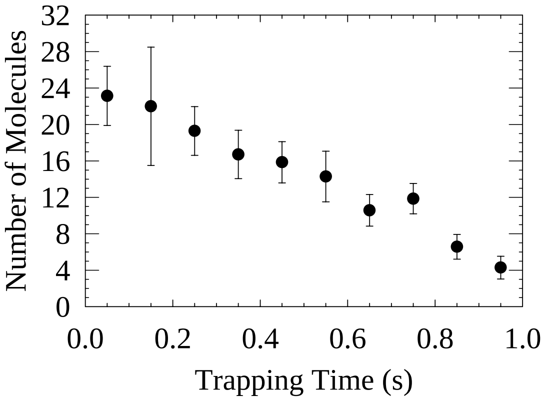

FIG. 3. Molecular trap lifetime (300 shots per point). The number of trapped molecules, as a function of time after the pushing laser is shuttered off. The pushing laser is shuttered on 50 ms after the MOT is turned off, and left on for 10 ms.

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