Two-photon photoionization of the Ca 4s3d $^{1}D_{2}$ level in an optical dipole trap

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We report an optical dipole trap for calcium. The trap is created by focusing a 488-nm argon-ion laser beam into a calcium magneto-optical trap. The argon-ion laser photoionizes atoms in the trap because of a near-resonance with the $4s4f^{1}F_{3}$ level. By measuring the dipole-trap decay rate as a function of argon-ion laser intensity, we determine the ${}^{1}F_{3}$ photoionization cross section at our wavelength to be approximately 230 Mb.

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I. INTRODUCTION

Laser-cooling experiments have expanded in recent years to group-II metals. This is partially due to a growing interest in optical frequency standards [1–3]. Calcium, strontium, and ytterbium all have narrow resonances from the ground state at relatively convenient laser wavelengths. The major isotopes of these elements have no angular momentum in the ground state, making the narrow "clock" transition frequencies less sensitive to external fields. Other experiments, such as metastable-collision studies, photoassociative spectroscopy and quasimolecule formation, Bose-Einstein condensation in simple atomic systems, and ultracold plasma investigations also contribute to the growing interest in lasercooling alkaline-earth metals.

The atomic density in dipole traps can be much higher than in magneto-optical traps. For some experiments, such as Bose-Einstein condensation, photoassociation, cold plasmas, and collision studies, higher densities can be helpful. Dipole traps for alkaline-earth atoms may also improve the performance of atomic clock experiments. For example, atoms can be held in a dipole trap generated by a laser at the so-called "magic wavelength" where the ac Stark shift is exactly equal for both atomic levels in the clock transition [4,5]. This would make it possible to use trapped neutral atoms for the clock, increasing the maximum interrogation time and therefore increasing the accuracy of the clock.

Only a few experiments have explored optical dipole traps for alkaline-earth atoms. The absence of angular momentum in the ground state prevents sub-Doppler cooling using resonance transitions [6], complicating dipole-trap loading. However, advanced cooling techniques can reduce the atomic temperature to a few microkelvin, and dipole traps in Sr [7,8] and Yb [9,10] have been reported in the literature. We are also aware of a ground-state calcium dipole trap reported in a Ph.D. thesis [11].

In this paper we report an optical dipole trap for calcium. Our trap captures $T \sim 1$ mK atoms in the excited $4s3d {}^{1}D_{2}$ metastable state. Because the atomic temperature is relatively high, the dipole trap operates in the regime where the light shift is several times larger than the natural atomic-transition linewidth [12]. The high intensity required to capture these atoms is also high enough to photoionize them. By measuring the photoionization production rate and the trap lifetime as a function of dipole-trap laser intensity, we determine the effective ${}^{1}D_{2}$ lifetime in our system and the photon-ionization cross section.

II. MAGNETO-OPTICAL TRAP

The calcium magneto-optical trap (MOT) is formed by three pairs of counterpropagating laser beams that intersect at right angles in the center of a magnetic quadrupole field [13]. The 423-nm laser light required for the calcium MOT is generated by frequency-doubling an infrared laser in KNbO₃, and has been described previously [14]. A diode laser master-oscillator-power-amplifier (MOPA) system delivers 300 mW single-frequency power at 846 nm, as shown in Fig. 1. This laser is phase locked to a buildup cavity using the Pound-Drever-Hall technique [15], giving a power enhancement of 30. A 10-mm-long a-cut KNbO₃ crystal in the small waist of the buildup cavity is used to generate 45 mW output power at 423 nm via noncritical phase matching at a temperature of $-12 \degree C$ [14].



FIG. 1. A schematic drawing of the MOT laser system and frequency-stabilization electronics used in these experiments.ACM: acousto-optical modulator. EOM: electro-optical modulator.

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FIG. 2. A partial energy-level diagram for calcium (not to scale).

The laser is further stabilized by locking the 423-nm light to the calcium resonance transition using saturated absorption spectroscopy in a calcium vapor cell [16]. An acoustooptic modulator (AOM) in one arm of the saturated absorption laser beams shifts the laser frequency so that the laser beam sent to the MOT is 35 MHz (one natural linewidth) below the atomic resonance. We also use the AOM to chop this beam and use a lock-in amplifier to eliminate the Doppler background in the saturated absorption signal. Because the 846-nm laser is already locked to the frequency-doubling cavity, the feedback from this second lock controls the frequency-doubling cavity length.

The trap is loaded from a thermal beam of calcium atoms that passes through the center of the MOT. The thermal beam is formed by heating calcium in a stainless steel oven to 650 °C. The beam is weakly collimated by the 1-mm-diameter, 10-mm-long aperture in the oven wall. As the beam passes through the MOT, the slowest atoms in the velocity distribution are cooled and trapped. An additional red-detuned (140 MHz, or four times the natural linewidth) laser beam counterpropagates the calcium atomic beam, significantly enhancing the MOT's capture efficiency. The density profile of the MOT is approximately Gaussian, with a $1/e^2$ radius of 0.5 mm and a peak density of 10^9 cm⁻³. The lifetime of the MOT is limited by optical pumping to the 4s3d ¹D₂ state (see Fig. 2).

III. OPTICAL DIPOLE TRAP

The dipole trap is formed by focusing a 488-nm argon-ion laser beam in the center of the MOT. The interaction of the laser beam with the atoms is easily described in terms of the ac-stark shift. The electric field of the laser beam \vec{E} induces a polarization \vec{P} in the atom. The interaction of these two fields gives rise to the $-\vec{P}\cdot\vec{E}$ potential. A rotating-wave approximation of this interaction leads to the well-known optical potential or "light shift"

$$U = \frac{\hbar \gamma^2}{8\Delta} \frac{I(r)}{I_s},\tag{1}$$

where \hbar is Planck's constant divided by 2π , $\gamma = \tau^{-1}$ is 2π times the natural linewidth, $I_s = \pi h c \gamma/3\lambda^3$ is the saturation

intensity, *c* is the speed of light, λ is the wavelength of the atomic transition, $\Delta = \omega - \omega_0$ is the detuning of the laser ω from the atomic transition ω_0 in rad/s, and I(r) is the intensity of the laser beam. For multilevel atoms with many transitions from a given state, the light-shift calculation in Eq. (1) is extended by summing contributions from all of the transitions connected to the level, replacing γ with the appropriate linewidths. We use the data tabulated in Refs. [17,18].

The 488-nm argon-ion laser wavelength is near-resonant with the $4s3d {}^{1}D_{2}-4s4f {}^{1}F_{3}$ transition [$\Delta = 2\pi c(1/487.9863 \text{ nm}-1/487.8126 \text{ nm})=-1.38 \times 10^{12}$]. For a 1-W laser beam focused to a 20- μ m Gaussian waist, the optical potential depth is $U/k_{B}=11.6$ mK. Such a deep potential is required to trap our relatively hot calcium atoms. The atomic temperature is near the Doppler limit of laser cooling using the 423-nm transition.

Dipole traps for heavier group-II atoms have been reported. Those experiments cooled on the intercombination lines, which have a much lower Doppler limit. This could be done in calcium, especially in conjunction with quenched cooling [19,20] or two-photon cooling [21]. However, no reports have been published to our knowledge.

IV. DIPOLE-TRAP LOADING

We load the dipole trap while the MOT light is on. The dipole trap fills up with atoms optically pumped into the ${}^{1}D_{2}$ state in the region of the 488-nm laser-beam focus. Other ${}^{1}D_{2}$ atoms from outside the focal region pass through the dipole trap, but there is no dissipative cooling mechanism to capture them.

Some of the ${}^{1}D_{2}$ atoms are ionized by the 488-nm laser beam via a two-photon transition to the continuum. This photoionization pathway is enhanced by a near resonance with the ${}^{1}F_{3}$ level. As discussed below, it is probably further enhanced by a near resonance in the Rydberg series leading up to the Ca II 3*d* ionization limit.

We measure the ion production rate for different 488-nm laser-beam intensities [22]. Sample data are plotted in Fig. 3(a). These data are replotted in Fig. 3(b) with the ion production rate divided by the laser power squared. This ratio is proportional to the number of ${}^{1}D_{2}$ atoms in the laser focus, and in the absence of a dipole trap, this signal should be a flat line. At low powers, the number of atoms in the dipole trap increases as the trap depth increases. The trap number maximizes when the average well depth is a few times the Doppler temperature. At higher power, the trap number falls off because the 488-nm laser beam shifts the $4s^{2} {}^{1}S_{0}$ and 4s4p $^{1}P_{1}$ levels out of resonance with the MOT laser, reducing the efficiency of the optical-pumping loading mechanism. We performed a Monte Carlo simulation of trap loading and found agreement with our data, as plotted in Fig. 3(b).

At higher laser powers, the ion production rate is somewhat higher than expected. This occurs when the light shift of the ${}^{1}S_{0}$ and ${}^{1}P_{1}$ levels due to the 488-nm laser beam exceeds the natural linewidth. This is precisely the condition under which ground-state atoms can be captured in the di-



FIG. 3. Ion count rate versus 488-nm laser power. (a) plots the raw count rate for a (measured) Gaussian waist of $w=20 \ \mu m$. The laser power was changed by varying the argon-ion laser tube current. The different symbols show measurements made with different intracavity laser apertures. (b) plots the ion count rate divided by the square of the laser power, which is proportional to the number of ${}^{1}D_{2}$ atoms in the dipole trap. The solid line is a fit of our Monte Carlo simulation of trap loading to the data.

pole trap. For these atoms the trap depth is comparable to the atom temperature (the Doppler cooling limit). Such an arrangement would increase the density of ground-state atoms, making the loading rate due to optical pumping higher at higher powers.

We can estimate the number of ${}^{1}D_{2}$ atoms in our dipole trap using a simple rate equation. For deep optical potentials in steady-state conditions, this is equal to the trap loading rate multiplied by the trap lifetime. The loading rate is equal to the optical pumping rate multiplied by the dipole-trap volume, and divided by the MOT volume. Because the confocal parameter of the 488-nm laser beam exceeds the MOT dimension, and because the dipole-trap oscillation period along the symmetry axis is long compared to the ${}^{1}D_{2}$ lifetime, we can assume that the volume ratio is just the square of the laser-beam waist divided by the square of the Gaussian size of the MOT cloud. The number of ${}^{1}D_{2}$ atoms in the dipole trap, N_{D} , can be written as

$$N_D = \frac{s/2}{1 + s + (2\Delta/\gamma)^2} \tau_{\rm eff} A N_S \left(\frac{w^2}{2r_0^2}\right),$$
 (2)

where $s=I/I_s$ is the saturation parameter, N_s is the number of ground-state atoms in the trap, $\tau_{\rm eff}$ is the dipole-trap lifetime, $A=2150 \ {\rm s}^{-1}$ is the Einstein A coefficient for the ${}^1P_1{}^{-1}D_2$ transition, and r_0 is the Gaussian $1/e^2$ radius of the MOT. Depending on the beam waist and laser power, this simple model tells us that we load up to 2000 atoms into the trap, for a peak density of approximately $5 \times 10^8 \ {\rm cm}^{-3}$. By comparison, the background density of 1D_2 atoms is given by Eq. (2) without the volume ratio, and with the numbers N_D and N_s replaced by densities. The 1D_2 background density is $\sim 10^7 \ {\rm cm}^{-3}$.

In view of these numbers, it is perhaps surprising that at the lowest laser powers we can detect a small number of atoms in the dipole trap, and discriminate against the background ${}^{1}D_{2}$ atoms. But the background atoms roll through the trap in a few microseconds, and the trapped atoms remain in the trap approximately 100 times longer. The photoionization probability increases with the time spent in the 488-nm laser focus, so the ionization signal is predominantly from the trapped atoms.

V. TWO-PHOTON PHOTOIONIZATION RATE

We measure the lifetime of the dipole trap by blocking the MOT laser beams, and measuring the decay of the ion signal. The three most important decay mechanisms are radiative decay of the ${}^{1}D_{2}$ level, collisional decay due to hot atoms from the thermal atomic beam, and two-photon ionization of the ${}^{1}D_{2}$ atoms. Because we do not have a suitable method for turning off the thermal beam, we cannot reliably extract the ${}^{1}D_{2}$ radiative lifetime. Published values of the lifetime are around 2 ms [23–27], somewhat longer than measured in our experiment. While our experiment cannot determine the radiative lifetime, by measuring the decay rate as a function of 488-nm laser intensity we can determine the photoionization cross section.

A rate equation for ${}^{1}D_{2}$ level decay in the dipole trap after the loading has turned off is

$$\frac{dN_D}{dt} = -N_D \left(\frac{1}{\tau_{\rm eff}} + \mathcal{A}I^2\right),\tag{3}$$

where \mathcal{A} is the two-photon ionization rate coefficient. This has the well-known solution

$$N_D(t) = N_D(0) \exp\left(\frac{1}{\tau_{\rm eff}} + \mathcal{A}I^2\right).$$
 (4)

In second-order perturbation theory, two-photon ionization is written as an overlap of the initial and final states summed over all possible intermediate states, divided by an energy denominator. For near-resonant ionization, the energy denominator makes the near-resonant term dominant, collapsing the sum to just one term. This one term looks like the product of the probability that an atom is excited into the ${}^{1}F_{3}$ state multiplied by the probability of photoionizing out of that state. We can write this term as

$$\mathcal{A}I^{2} = \frac{s/2}{1+s+(2\Delta/\gamma)^{2}}\frac{I}{h\nu}\sigma$$
(5)

$$=\frac{3\lambda^4\sigma\gamma}{8\pi\hbar^2c^2\Delta^2}l^2,$$
(6)

where σ is the ${}^{1}F_{3}$ one-photon photoionization cross section. The approximation in Eq. (6) assumes that $s \ll (2\pi\Delta/\gamma)^{2}$ and $\gamma \ll \Delta$. In our experiment, the intensity of the 488-nm laser has a Gaussian spatial profile. Averaging the square of the intensity over the laser profile allows us to relate the photoionization rate to the total laser power. In this case, it can be written as

$$\mathcal{A}I^2 = \frac{\lambda^4 \sigma \gamma}{2\pi^3 h^2 c^2 \Delta^2 w^4} P^2. \tag{7}$$



FIG. 4. Dipole-trap decay versus power squared. The decay signal from the dipole trap. (a) shows the ion signal versus time with 1 W focused to a 90- μ m waist. The effective decay rate for these data is 1.02 kHz. (b) shows the apparent decay rate for a range of powers, all focused to a 90- μ m waist. The decay rate is fitted to a line proportional to the square of the power, as suggested by Eq. (3).

The dipole-trap decay rate as a function of the square of the 488-nm laser power is shown in Fig. 4. For each power level, we measured the dipole-trap decay. At sufficiently low power levels, this decay is approximately exponential, and we extract the decay rate using a least-squares fitting routine [28]. The decay rate depends on power. The zeroextrapolated decay rate is $\tau_{\rm eff}^{-1}$ =0.93 kHz. This rate is approximately twice the radiative decay rate. We see evidence in our experiment that our rate is significantly influenced by collisions with atoms in the thermal atomic beam. This is not surprising because the dipole trap sits in the middle of the thermal atomic beam. Without detailed characterization of the thermal beam, we cannot reliably extract a thermalatom-1D₂-atom collision cross section.

The slope of the decay rate with power is 80 Hz/W^2 . Using Eq. (7), we can determine the 1F_3 photoionization cross section. This gives a photoionization cross section for the ${}^{1}F_{3}$ level of $\sigma = 230 \times 10^{-18}$ cm². This extraordinarily large cross section suggests that the final state lies near a Rydberg state in the continuum. The final state is 133 cm⁻¹ below the Ca II ${}^{2}D_{3/2}$ ionization limit. The principal quantum number of hydrogenic Rydberg levels in this region is $n \sim 29$, and the separation between levels is 9 cm⁻¹. Our measurements are carried out in the presence of an electric field, further increasing the probability of finding a nearby Rydberg level. The NIST database tabulates only odd-parity levels in this energy region. We are unaware of applicable quantum defect calculations or measurements.

VI. CONCLUSION

We have demonstrated an optical dipole trap for neutral calcium atoms. These atoms are nonadiabatically loaded into the trap by an optical pumping mechanism. The lifetime of our trap is limited by the ~ 2 ms lifetime of the ${}^{1}D_{2}$ atoms and by collisions with atoms in the thermal atomic beam.

Our initial interest in the calcium optical dipole trap was its potential application to our ultracold plasma research. It may be possible to use the dipole trap as the beginning point for ultracold plasma expansion studies. Because of the high aspect ratio, a plasma generated from a dipole trap would be a two-dimensional ultracold neutral plasma at early times. Correlation heating would be reduced compared to the threedimensional case [29], bringing the two-dimensional plasma closer to the strongly coupled regime. Furthermore, the plasma expansion depends on the exact density distribution of the initial cloud. Compared to standard MOT traps, a dipole trap has a well-defined density profile, meaning that these new plasmas could greatly improve the reproducibility of plasma expansion experiments. We plan to explore these possibilities in future work.

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