

Photoassociative Spectroscopy at Long Range in Ultracold Strontium

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(Received 5 October 2004; revised manuscript received 18 February 2005; published 3 March 2005)

We report photoassociative spectroscopy of $^{88}\text{Sr}_2$ in a magneto-optical trap operating on the $^1S_0 \rightarrow ^3P_1$ intercombination line at 689 nm. Photoassociative transitions are driven with a laser red detuned by 600–2400 MHz from the $^1S_0 \rightarrow ^1P_1$ atomic resonance at 461 nm. Photoassociation takes place at extremely large internuclear separation, and the photoassociative spectrum is strongly affected by relativistic retardation. A fit of the transition frequencies determines the 1P_1 atomic lifetime ($\tau = 5.22 \pm 0.03$ ns) and resolves a discrepancy between experiment and recent theoretical calculations.

DOI: 10.1103/PhysRevLett.94.083004

PACS numbers: 32.80.Pj

Photoassociative spectroscopy (PAS) in laser-cooled gases is a powerful probe of molecular potentials and atomic cold collisions. It provides an accurate determination of ground state scattering lengths and excited state lifetimes [1]. Photoassociation occurs naturally in laser cooling and trapping experiments in which the lasers are red detuned from atomic resonance, so characterizing the process is important for understanding and optimizing the production of ultracold atoms (e.g., [2]). PAS is also used to produce ultracold molecules [3] and to manipulate scattering lengths [4].

PAS of alkaline earth atoms differs significantly from more common studies of alkali metal atoms. The most abundant isotopes of alkaline earth atoms lack hyperfine structure, making these systems ideal for testing PAS theory. Recent theoretical work [5] emphasized the ability to resolve transitions at extremely large internuclear separation and very small detuning from the atomic asymptote. The finite speed of light modifies the potential in this regime through relativistic retardation [6–8].

There is also interest in cold collisions of alkaline earth atoms and atoms with similar structure because of their importance for optical frequency standards [9–12] and for the creation of quantum degenerate gases [13–15]. In addition, collisions involving metastable states [16–19] display novel properties arising from electric quadrupole-quadrupole or magnetic dipole-dipole interactions.

PAS red detuned from the principal transitions in calcium [20] and ytterbium [21] is well characterized, resulting in accurate measurements of the first excited 1P_1 state lifetimes and the ground state s -wave scattering lengths. In spite of its importance for potential optical frequency standards, little is known about strontium. The photoassociative loss rate induced by trap lasers in a $^1S_0 \rightarrow ^1P_1$ magneto-optical trap (MOT) has been measured [2]. *Ab initio* strontium potentials have been calculated for small internuclear separation ($R < 1$ nm) [22].

In this Letter we report PAS of ^{88}Sr near the $^1S_0 \rightarrow ^1P_1$ atomic resonance at 461 nm (Fig. 1). The simple spectrum allows us to resolve transitions as little as 600 MHz de-

tuned from the atomic resonance, which produces molecules with very large internuclear separation. Our determination of the first excited 1P_1 lifetime resolves a discrepancy between experiment and recent theoretical work, and provides an importance test of atomic structure theory for alkaline earth atoms.

The most common form of PAS involves resonantly inducing trap loss in a MOT, although early work was also conducted in optical-dipole traps [1]. Recent experiments also studied spectroscopy of atoms in magnetic traps, especially in Bose-Einstein condensates [23,24]. The experiments described in this Letter were performed in a MOT, but the MOT operated on the $^1S_0 \rightarrow ^3P_1$ intercombination line at 689 nm [25], rather than an electric-dipole allowed transition. This results in lower atom temperature, a shallower trap, and higher atom density. The signals are

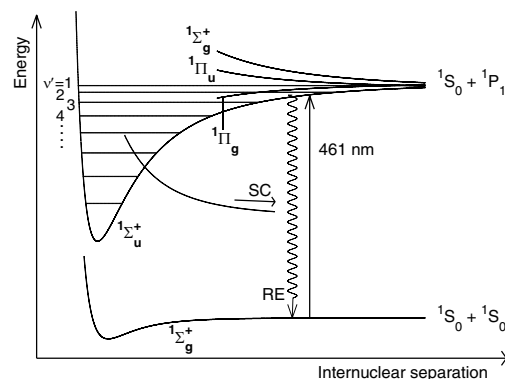


FIG. 1. Molecular levels and photoassociative transitions. Resonant excitation occurs near the outer molecular turning point of states of the $^1\Sigma_u^+$ potential, and it can lead to trap loss through two processes. The molecule can radiatively decay at smaller internuclear separation to two ground state atoms with kinetic energies exceeding the trap depth. This is known as radiative escape (RE). In a state-changing collision (SC), at small internuclear separation the molecular state changes to one corresponding at long range to a lower-lying electronic configuration of free atoms. The atoms exit the collision with greatly increased kinetic energy and escape the trap.

strong enough that we can obtain PAS spectra with very low PAS laser intensity. This avoids many complications found or predicted in high intensity spectra [26,27].

Atoms are initially trapped in a MOT operating on the 461 nm $^1S_0 \rightarrow ^1P_1$ transition, as described in [28]. During the loading phase, the peak intensity in each MOT beam is 6 mW/cm², and the axial magnetic field gradient generated by a pair of anti-Helmholtz coils is 56 G/cm. The intensity is then reduced by about a factor of 8 for 3.5 ms to reduce the atom temperature. After this stage the MOT contains about 50×10^6 atoms at 2 mK.

The 461 nm laser cooling light is then extinguished, the field gradient is reduced to 2.1 G/cm, and the 689 nm light for the $^1S_0 \rightarrow ^3P_1$ intercombination line MOT is switched on. This MOT also consists of three retro-reflected beams, each with a diameter of 2 cm and intensity of 400 μ W/cm². The frequency of the 689 nm laser is detuned from the atomic resonance by 0.5 MHz and spectrally broadened with a ± 400 kHz sine-wave modulation. Transfer and equilibration take about 50 ms, after which there are 15×10^6 atoms at a temperature of 5 μ K, peak density of about 5×10^{11} cm⁻³, and $1/\sqrt{e}$ density radius of about 100 μ m.

The intercombination line MOT operates for an adjustable hold time before measuring the remaining number of atoms with absorption imaging using the $^1S_0 \rightarrow ^1P_1$ transition. The lifetime of atoms in the MOT is approximately 350 ms, limited by background gas collisions. To detect photoassociative resonances, a PAS laser is applied to the atoms during hold times of 300–400 ms. When the PAS laser is tuned to a molecular resonance, photoassociation provides another loss mechanism for the MOT, decreasing the number of atoms.

Light for photoassociation is derived from the same laser that produces the 461 nm light for laser cooling. Several acousto-optic modulators (AOM's) detune the light 600 to 2400 MHz to the red of the atomic transition. The laser frequency is locked relative to a Doppler-free saturated-absorption feature in a vapor cell, with an uncertainty of about 2 MHz. The last AOM in the offset chain, in a double-pass configuration, scans the frequency up to 200 MHz with minimal beam misalignment. We adjust the rf power to the AOM to maintain the PAS laser intensity at a constant level during any single scan. PAS light is double-passed through the MOT in a standing wave, with a $1/e^2$ intensity radius of $w = 3$ mm.

Figure 2(a) shows the number of atoms remaining after the hold time as a function of PAS laser detuning near a typical resonance. We record spectra with different PAS laser intensities, ranging from 0.5–27 mW/cm². (The atomic saturation intensity is $I_{\text{sat}} = 40$ mW/cm².) For the lower intensities, the linewidth is close to the expected natural linewidth of twice the inverse of the 1P_1 atomic lifetime ($\tau^{-1} = 2\pi \times 30.5$ MHz). Thermal broadening ($k_B T/h \approx 100$ kHz) is negligible, as are light, polarization, Zeeman, and predissociation shifts. Only s -wave

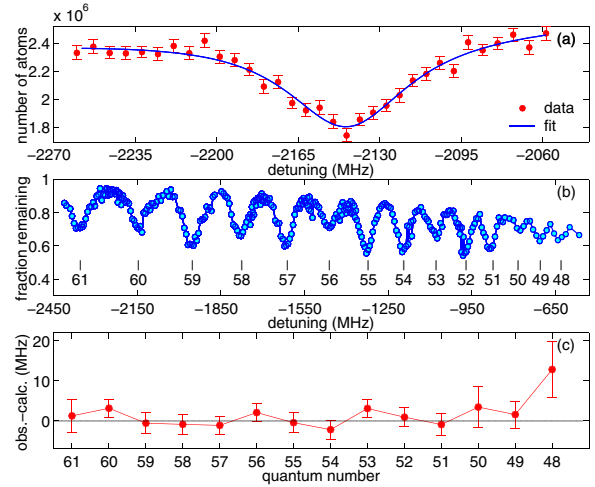


FIG. 2 (color online). (a) Spectrum for photoassociation to the $v' = 60$ molecular state. Detuning is with respect to the atomic $^1S_0 - ^1P_1$ resonance. The PAS laser intensity is 1.4 mW/cm². The Lorentzian fit yields a FWHM linewidth of 70 ± 7 MHz. (b) Representative spectra for all transitions observed in this study. Lines connect the data points to guide the eye. Conditions vary for the individual scans that make up the full spectrum, so amplitudes and linewidths should not be quantitatively compared. The baseline has been adjusted to match the expected curve for overlapping Lorentzians. (c) Differences between experimental and calculated positions of the 14 measured levels.

collisions occur, so only a single rotational level ($J = 1$) is excited. For higher intensity, the observed line is broadened slightly because, on resonance, the signal saturates if a large fraction of atoms is lost.

Figure 2(b) shows a complete spectrum of the 14 different PAS resonances observed in this study. Center frequencies for each transition are determined by Lorentzian fits. The typical statistical uncertainty is 2–3 MHz, but there is approximately 2 MHz additional uncertainty arising from the lock of the laser. The Condon radius for the excitation varies from 20 nm for the largest detuning to 32 nm for the smallest. These extremely large values are comparable to the internuclear spacing of molecules formed in photoassociative spectroscopy of pure long-range potentials [7,29,30]. For smallest detuning, the linewidth is comparable to the level spacing. This corresponds to the classical situation of the lifetime being on the order of the vibrational period.

The region of the attractive $^1\Sigma_u^+$ molecular potential probed by PAS corresponds to large internuclear separations and is typically approximated by

$$V(R) = D - \frac{C_3}{R^3} + \frac{\hbar^2[J(J+1) + 2]}{2\mu R^2}, \quad C_3 = \frac{3\hbar\lambda^3}{16\pi^3\tau}, \quad (1)$$

where D is the dissociation energy, μ the reduced mass, and $\lambda = 461$ nm. However, at very large separations, the atom-atom interaction is modified by relativistic corrections. Retardation effects [6] can be included in the analy-

sis of the spectrum through $C_3 \rightarrow C_3[\cos(u) + u \sin(u)]$, where $u = 2\pi R/\lambda$. Machholm *et al.* [5] discussed this in the context of PAS of alkaline earth atoms.

To extract molecular parameters from the positions of the PAS resonances, we constructed a potential curve by smoothly connecting the long-range form [Eq. (1)] to a short range curve at a distance of 1.5 nm. The short range *ab initio* potential was obtained using a semiempirical two-valence-electron pseudopotential method [22]. To account for uncertainty in the short range potential, the position of the repulsive wall was varied as a fit parameter. The rovibrational levels in the $^1\Sigma_u^+$ potential were found by solving the radial Schrödinger equation using the mapped Fourier grid method [31] with a grid size typically larger than 500 nm containing about 1000 grid points.

We found that the observed levels range from $v' = 48$ to 61, where we count levels from the dissociation limit down as $v' = 1, 2, \dots$, and $v_D - v = v' - 1 + (v_D \text{ modulo } 1)$, where v is the standard vibrational level label and v_D is the noninteger number corresponding to a hypothetical level at the dissociation energy [20]. The best fit of the data was achieved with $C_3 = 18.54$ a.u., with a reduced chi-squared value of $\chi^2 = 0.79$. [See Fig. 2(c)]. We have a 1-standard-deviation uncertainty of 1 in the level assignment because χ^2 increased by 8% for a change of level assignments of $\Delta v' = \pm 1$. The value of C_3 changed by $\pm 0.5\%$ as the assignments changed by ± 1 , which is much larger than the statistical uncertainty in C_3 for a given assignment. The χ^2 contours thus imply a 1-standard-deviation uncertainty in C_3 of 0.5%. A fit to the level spacings, as opposed to the absolute positions, yielded the same value of C_3 . From C_3 , we derive a natural decay rate of the atomic $5p\ ^1P_1$ state of $\tau = 5.22 \pm 0.03$ ns. Decay channels other than $^1P_1 \rightarrow ^1S_0$ can be neglected at this level of accuracy. The most recent experimental determinations of τ use the Hanle effect [32,33]. Our result agrees well with recent theoretical values [34–36] (Fig. 3).

Retardation effects shift the levels by approximately 100 MHz in this regime, which is similar to the shift seen in a pure long-range potential in Na_2 [7]. If retardation effects are neglected and the data is fit using the simple semiclassical treatment [37], the level assignments change significantly, and C_3 changes by more than 10%, putting it outside the range of recent theoretical results. Center fre-

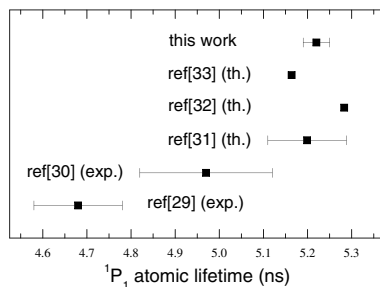


FIG. 3. Comparison of experimental (exp.) and theoretical (th.) values for the lifetime of the 1P_1 level.

quency shifts due to coupling to the continuum of the ground state have been ignored as they are less than 50 kHz for typical experimental temperatures.

Figure 4(a) shows an example of the number of atoms, N , in the intercombination line MOT as a function of time. The density in the MOT varies according to $\dot{n} = -\Gamma n - \beta n^2$. Integrating this equation over volume and solving the differential equation yields

$$N(t) = N_0 \frac{e^{-\Gamma t}}{1 + \frac{N_0 \beta}{\Gamma(\sqrt{4\pi\sigma})^3} (1 - e^{-\Gamma t})}. \quad (2)$$

We approximate the density as $n(r) = n_0 e^{-r^2/2\sigma^2}$, which is appropriate because the cloud is not optically thick to MOT photons. Γ is the one-body loss due primarily to background gas collisions. β is the two-body loss rate and it contains important information about the dynamics of photoassociation and trap loss.

Even with photoassociation, the deviation of $N(t)$ from a single exponential decay is small, making it difficult to independently extract β and Γ with high accuracy from a single decay curve. To address this problem, we take advantage of the fact that when the PAS laser is not on a molecular resonance, the photoassociation rate is small [See Fig. 2(b)], and within the accuracy of our measurement it can be neglected. All other processes, however, are the same. We thus take data in pairs of on and off resonance, and fit off-resonance data to $N(t) = N_0 e^{-\Gamma t}$. The on-resonance data is then fit with Eq. (2) with Γ fixed to the value determined from the off-resonance data. The resulting two-body decay rates are shown in Fig. 4(b). For the relatively small intensities used, β is expected to vary linearly with laser power [38].

We currently image along the direction of gravity and lack the additional diagnostic required to obtain information on the third dimension of the atom cloud. Gravity can distort the equilibrium shape of the intercombination line MOT because the light force is so weak. However, we operate the MOT in the regime where the laser detuning

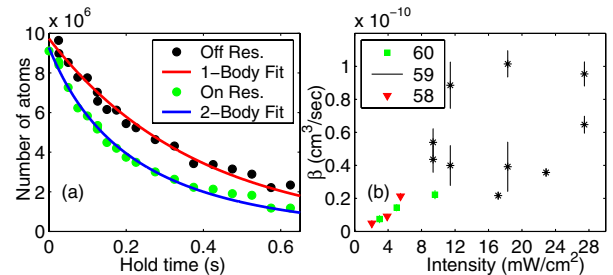


FIG. 4 (color online). (a) Number of atoms as a function of time for atoms in the intercombination line MOT. The PAS laser is tuned near the $v' = 59$ molecular resonance. In one data set the PAS laser is on resonance, and in the other it is tuned to the blue of resonance by 42 MHz, and loss due to photoassociation is small. (b) β as a function of PAS laser intensity for several molecular resonances. Error bars denote statistical uncertainties.

is comparable to the modulation of the laser spectrum, and, as shown in [39], in this regime the effect is small. Based on the range of distortions we observe in the imaged dimensions for various misalignments of the MOT beams, we allow for a scale uncertainty of a factor of 2 in the volume of the MOT. This contributes an identical uncertainty in the measurements of β .

Measurements of MOT size suggest that as the PAS laser power increases, the cloud radius increases slightly (20% for the highest intensities), decreasing the atomic density. This is expected because off-resonance scattering from the atomic transition should heat the sample. The data quality does not allow a reliable correction for this effect, however, and we use the average of all observed σ as a constant σ in Eq. (2). This contributes another approximately 50% uncertainty in our determination of β . Overall, we quote a factor of 3 uncertainty due to systematic effects, which dominates over statistical error.

The measured values of $\beta \approx 4 \times 10^{-12}$ cm³/sec for 1 mW/cm² in the intercombination line MOT are comparable to theoretical [5] values of β for atoms in a MOT based on an electric-dipole allowed transition, which would typically have a trap depth of about 1 K. In our experiment, the transfer efficiency of atoms from the 461 nm MOT places a lower limit of 0.5 mK on the depth of the intercombination line MOT. Using the formalism of [39] and the parameters of the intercombination line MOT, we calculate a trap depth of 2 mK. As described in Fig. 1, photoassociative loss requires excitation to a molecular state followed by decay to a configuration of two atoms with enough kinetic energy to escape from the trap [5]. The fact that the observed linewidths are close to the theoretical minimum of $1/(\pi\tau)$ suggests that radiative escape dominates over state-changing collisions in this regime, as predicted in theoretical calculations of PAS rates for magnesium [5]. If β is similar for trap depths of 1 K and 1 mK, then the energy released during radiative decay must be on the order of 1 K or greater. Extensive calculations of radiative escape in lithium [40] also led to an estimate of a few kelvin for the energy released in this process.

Photoassociative spectroscopy has probed dynamics of collisions in an intercombination line MOT and provided an accurate measurement of the lifetime of the lowest 1P_1 state of strontium. This has resolved a previous discrepancy between experimental and theoretical values of the lifetime. We have also taken advantage of the simple structure of the spectrum to measure transitions at very large internuclear separation where the level spacing and natural linewidth are comparable and retardation effects are large.

This research was supported by the Office for Naval Research, National Science Foundation, Welch Foundation, Research Corporation, Alfred P. Sloan Foundation, and David and Lucille Packard Foundation. The authors are grateful to A. R. Allouche for providing them the Sr₂ *ab initio* potentials.

Note added.—Since the submission of this manuscript, results of PAS of ⁸⁸Sr₂ were posted [41] that yield a C₃

coefficient in reasonable agreement with the value presented here and a calculation of the *s*-wave scattering length.

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