$7S_{1/2} \rightarrow 9S_{1/2}$ two-photon spectroscopy of trapped francium

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Received August 13, 1996

We report on the spectroscopic measurement of the ^{210}Fr $9S_{1/2}$ energy obtained by two-photon excitation of atoms confined and cooled in a magneto-optic trap. The resonant intermediate level $7P_{3/2}$ is the upper state of the trapping transition. We have measured the energy difference between the $9S_{1/2}$ state and the $7S_{1/2}$ ground state to be 25 671.021 ± 0.006 cm^{-1}. © 1996 Optical Society of America

Francium (Fr) is the heaviest of the alkali atoms and has no stable isotopes. It occurs naturally from the α decay of actinium or artificially from fusion or spallation nuclear reactions in an accelerator. Its longest-lived isotope has a half-life of 21.8 min. In previous experiments the study of the atomic structure of Fr was possible only with the very high fluxes available at a few facilities in the world[1] or by use of natural sources.[2] We have recently captured Fr atoms[3] in a magneto-optic trap (MOT), opening the possibility for extensive studies of its atomic structure.

The motivation for this program is the study of the fundamental interactions in atoms. The simple structure of alkali atoms permits precise calculations, which make them useful for this purpose. Fr, as the heaviest alkali, offers the possibility of a most sensitive test of the standard model through an atomic parity nonconservation measurement.[4] It is, however, the least studied of the alkalis.

We have studied the atomic properties of Fr in a MOT on line with an accelerator. The captured atoms are confined for long periods of time in a small volume moving at low velocity, an ideal environment for precision spectroscopy.[5–8] In this Letter we present our measurement of the $7S_{1/2} \rightarrow 9S_{1/2}$ energy interval by resonant two-photon spectroscopy.

A diagram of the apparatus is shown in Fig. 1. A beam of 6.3×10^{11} ^{18}O ions/s from the Stony Brook superconducting linear accelerator (LINAC) impinging upon a Au target produces $\approx 1 \times 10^6$/s ^{210}Fr in the target that has a half-life of 3.2 min. We separate the production and the trapping regions by extracting the Fr atoms out of the Au as ions and transport them 1 m, where they are neutralized upon a heated yttrium surface. The neutral atoms are released into a nonstick dry-film-coated glass cell where the MOT forms.

The physical trap consists of a 10-cm-diameter Pyrex bulb with six 5-cm-diameter windows and two 3-cm-diameter viewing windows. The MOT is formed by six intersecting laser beams, each with a $1/e^2$ (power) diameter of 4 cm and a power of 50 mW, with a magnetic field gradient of 9 G/cm. The $1/e$ fill time of the trap is 20 s.

The trap operates on line in the target room of the accelerator, and we remotely control the experiment. Before trapping Fr, we tested the apparatus extensively by injecting stable ^{87}Rb into the cell. The Rb comes from a dispenser next to the Au target and follows the same path through the apparatus as the Fr. We use the trapped Rb to optimize the position of the photomultipliers that detect the trapped atoms. Then we change the laser frequencies to the Fr resonances without further adjustment of the detection optics.

The energy levels of the ^{210}Fr atom relevant for trapping and for the two-photon spectroscopy are shown in Fig. 2. A Coherent 899-21 Ti:sapphire laser with a rms linewidth of less than 500 kHz excites the cycling transition $7S_{1/2}$, $F = 13/2 \rightarrow 7P_{3/2}$, $F = 15/2$ at 718 nm. Atoms that fall into the $7S_{1/2}$, $F = 11/2$ ground state 46.8 GHz lower than the $F = 13/2$ state return to the cycling transition with a repumper laser. The repumper laser is a 1-mW free-running Mitsubishi 4405-01 diode laser cooled to approximately 77 K, modulated in frequency by 500 MHz with a rate of 10 kHz. The frequency of the laser is monitored with an I_2 absorption cell in the region between lines 387 and 388.[9]

To detect the 718-nm fluorescence from the captured atoms, we frequency modulate the trapping laser with an amplitude of 4.7 MHz at a rate of 14.5 kHz. Lock-in detection permits the rejection of the laser light

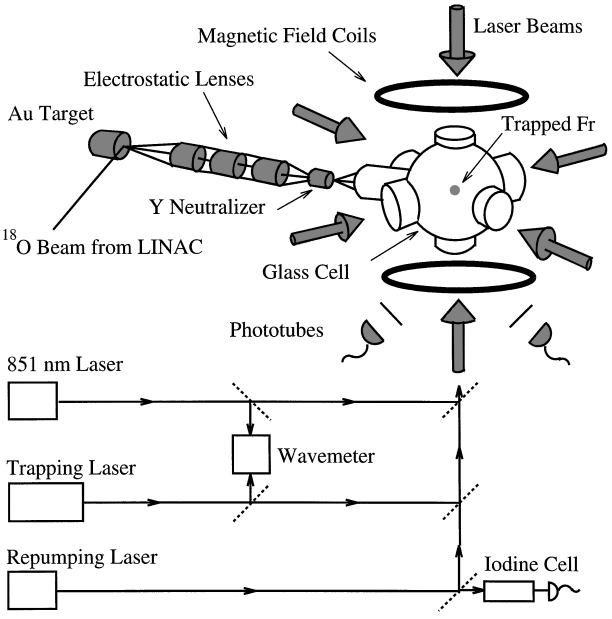

Fig. 1. Diagram of the experimental setup.

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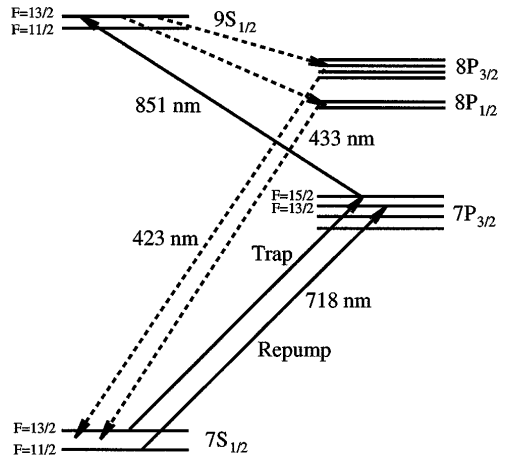

Fig. 2. Diagram of energy levels of ^{210}Fr that are relevant for trapping and two-photon excitation.

scattered from the cell while we are measuring the fluorescence from the captured atoms. An $f/2$ optical system collects the trap fluorescence in a Hamamatsu R636-10 photomultiplier tube, and the signal is demodulated by a Stanford Research SR830 lock-in detector. In addition, two cameras view the region of the trap to monitor the fluorescence and the shape of the trap.

In a previous measurement[3] the frequency of the Ti:sapphire laser was referenced to the tabulated line 381 of the atlas of the I_2 spectrum.[9] This allowed us to determine the frequency of the trap resonance as $13\,923.3866 \pm 0.0026\ \mathrm{cm}^{-1}$. The width of the trap resonance is 10 MHz.

We first scan the frequency of the trapping laser and look for a signal coming from the 718-nm fluorescence in the output of the lock-in amplifier. A small portion of the trapping laser light goes into a Burleigh WA-1500 wavemeter, which determines the wavelength to a fractional precision of 10^{-7}. When the trap forms, we stop the scan and view the fluorescent Fr atoms on a TV monitor. We manually adjust the Ti:sapphire frequency (stability $\approx$50 MHz/h) to account for drifts of the trap laser out of the trap resonance, guided by the change in shape of the cloud of trapped atoms.

An additional laser excites the transition $7P_{3/2}$, $F = 15/2 \rightarrow 9S_{1/2}$, $F = 13/2$. This laser is an Environmental Optical Sensors, Inc. (EOSI) 2001 external-cavity diode laser operating at 851 nm, which is locked to a thermally stabilized étalon with broadband mirrors and a 500-MHz free-spectral range. To scan the frequency of the 851-nm laser we scan the étalon and the laser follows, which permits slow scans. We found this procedure necessary to reduce the long-term excursions (seconds to minutes) of the laser frequency. The resulting laser linewidth of greater than a minute as measured by an independent Fabry–Perot interferometer is less than 10 MHz.

The *ab initio* calculations by Dzuba *et al.*[10] of the ionization energy of the $9S_{1/2}$ state have an upper-limit accuracy of 0.3%. A search over such a large range is difficult, so we used quantum defect calculations by Bergeman[11] and Kim[12] as a guide. Their empirical calculations incorporated the ionization potential as well as the location of the levels from $10S$ to $22S$ from Andreev *et al.*[2] and the extensive research effort carried out at the ISOLDE.[1] The results of these studies allowed us to search for the $9S_{1/2}$ state with a window of 1 cm^{-1} around the quantum defect prediction.

With a cloud of more than 10^4 ^{210}Fr atoms we start a scan of the EOSI laser at a rate of 0.7 MHz/s. A portion of this laser beam also enters the wavemeter and is read alternately with the trapping laser (see Fig. 1).

We identify the transition in two ways. We detect the fluorescence of the atoms that have been excited to the $9S_{1/2}$, $F = 13/2$ state as they decay back to the $7S_{1/2}$ ground state by the $8P_{3/2}$ and $8P_{1/2}$ states and emit photons at 423 and 433 nm. These photons pass through colored glass and interference filters and are detected with a blue-sensitive photomultiplier Amperex XP 2020Q [Fig. 3(a)]. There is a large background of $\sim$1500 counts/s in the photon-counting trace coming from the nuclear reactions taking place less than 2 m away from the photomultiplier. The 718-nm trap fluorescence is also a sensitive indicator of the transition. As trapped atoms undergo the $7P_{3/2} \rightarrow 9S_{1/2}$ transition they can decay by a path other than the $7P_{3/2}$ state. This reduces the signal of the lock-in amplifier that monitors the 718-nm fluorescence from the cycling transition [Fig. 3(b)]; cf. Ref. 8. The asymmetry seen in both traces reflects the Autler–Townes splitting of the $7P_{3/2}$, $F = 15/2$ state. The wave number is measured at the peak of the resonance with the Burleigh wavemeter. The peak corresponds to the coherent addition of the two photons.[13]

We take the ratio R between wave numbers of the $7P_{3/2}$, $F = 15/2 \rightarrow 9S_{1/2}$, $F = 13/2$ transition to the

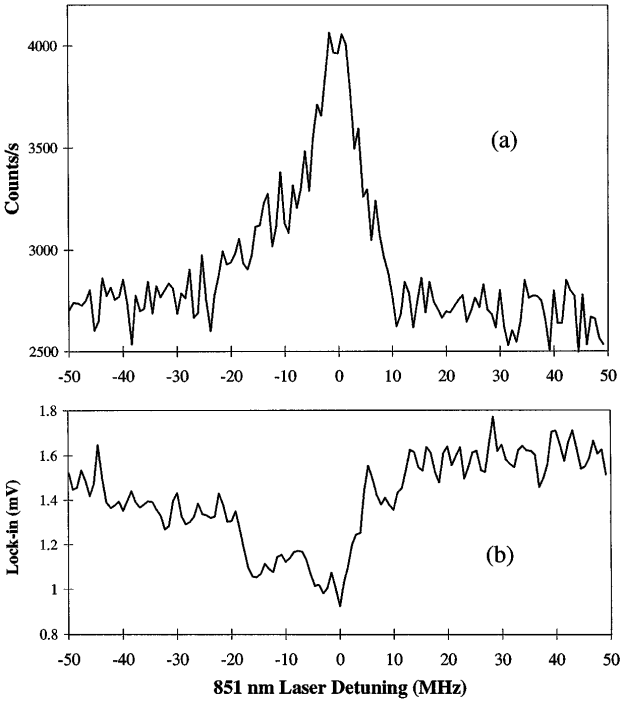

Fig. 3. (a) Fluorescence at 423 and 433 nm versus 851 nm laser frequency detuning (100-MHz scan). (b) Trap fluorescence at 718 nm versus 851-nm laser frequency detuning (100-MHz scan). The zero of the scan corresponds to $11\,746.978 \pm 0.003\ \mathrm{cm}^{-1}$.

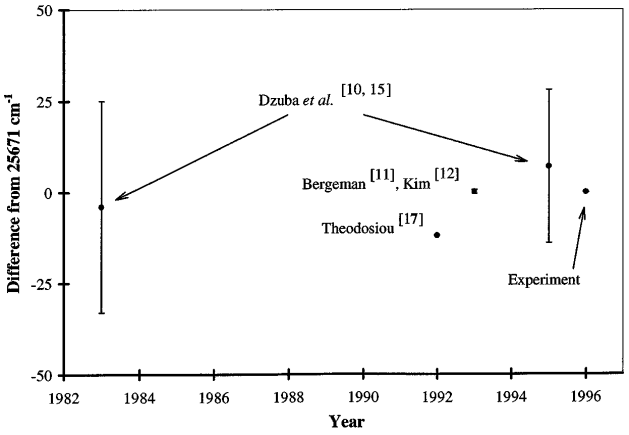

Fig. 4. Comparison of *ab initio* predictions of Refs. 10 and 15 and quantum defect interpolations of Refs. 11, 12, and 17 with the experimental measurement of the $7S_{1/2}$–$9S_{1/2}$ energy interval.

$7S_{1/2}, F = 13/2 \rightarrow 7P_{3/2}, F = 15/2$ transition measured with the wavemeter and use our absolute measurement of the trap resonance[3] to reduce systematic errors with the wavemeter. From scans with a FWHM of 15 MHz like the one shown in Fig. 3 we have determined that the ratio $R = 0.84368685 \pm 1.1 \times 10^{-7}$.

Although there is a small population in the $7P_{3/2}$, $F = 13/2$ by way of the repumping laser it is not enough to give a significant signal for the $F = 11/2$ hyperfine level of the $9S_{1/2}$ state. We calculate the hyperfine shift of the $9S_{1/2}$, $F = 13/2$ state (1920 ± 58 MHz) based on the research of Arnold *et al.*[1] and Coc *et al.*[14] and use the measurement of the $7S_{1/2}$, $F = 13/2$ ground-state hyperfine shift (21 585.3 ± 1.2 MHz) (Ref. 14) to find the center of gravity of the transition.

From the images of the captured atoms the size of the trap is less than 1 mm in diameter. The magnetic field cannot be zero in the place where the trap forms and can cause a shift in the frequency that we estimate to be smaller than the linewidth. Scans were performed with increasing and decreasing frequency with a repeatability of better than 0.001 cm^{-1}. The linewidth of the transition is narrow, and our precision is limited by the I_2 reference and the wavemeter. The center-of-gravity energy interval between the $7S_{1/2} \rightarrow 9S_{1/2}$ states of ^{210}Fr is 25 671.021 ± 0.006 cm^{-1}.

Figure 4 shows the comparison of our measurement with *ab initio* calculations of Dzuba *et al.*[10,15,16] as well as with three semiempirical quantum defect fits.[11,12,17] The calculations of Bergeman and Kim give the closest agreement with the measurement, but it is impressive to see the excellent prediction of the *ab initio* calculation.

We have performed two-photon spectroscopy of francium in a MOT, using the cycling transition of the trap as the intermediate resonant step, and have determined the energy interval between the $7S_{1/2}$ and $9S_{1/2}$ states. The location of this state is necessary for the continuing effort to study parity nonconservation in francium.

We thank T. H. Bergeman and Y.-K. Kim for their quantum defect calculations and the Northrop Grumman Research Center for extended loans of imaging equipment. This study was supported in part by a Precision Measurement Grant from the National Institute of Standards and Technology and by the National Science Foundation.

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