Two-photon spectroscopy of the francium $8S_{1/2}$ level


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We report the spectroscopy of the $8S_{1/2}$ energy level in $^{210}$Fr by two-photon excitation of atoms confined and cooled in a magneto-optical trap. The resonant intermediate level is the upper state of either the $7P_{3/2}$ trapping transition or the $7P_{1/2}$ repumping transition. The center-of-gravity energy difference between the $7S_{1/2}$ ground level and the $8S_{1/2}$ level is $19732.523\pm0.004$ cm$^{-1}$. The hyperfine separation of the $8S_{1/2}, F=11/2$ and $F=13/2$ states is $10.256\pm7$ MHz, giving a magnetic dipole hyperfine constant $A$ of $1577.8\pm1.1$ MHz. [S1050-2947(99)01201-9]

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Francium has the simplest electronic structure of the heavy elements but has no stable isotopes. Spectroscopic measurements of the Fr atomic structure test ab initio calculations that predict energy levels and hyperfine splittings [1–4]. In particular, the hyperfine structure is sensitive to relativistic, core polarization, and core correlation effects that are difficult to calculate accurately in multielectron atoms. A further motivation for its study is the possibility of performing spectroscopic measurements that test fundamental symmetries in Fr.

While the atomic structure of most alkali-metal atoms has been extensively studied, there is no similar knowledge for Fr. Many levels and hyperfine splittings were located in the experiments at ISOLDE [5] and with a natural source [6]. It is now possible to study francium in an ideal environment for spectroscopy: a magneto-optical trap on-line with an accelerator that produces it [7] or with a natural source [8]. Recently measured atomic properties include the location of the $9S_{1/2}$ level [9], the lifetimes of the $7P_{3/2}$ and $7P_{1/2}$ levels [10,11] and accurate measurements of the hyperfine splittings of the $7p$ levels [8].

A better understanding of the complex francium atom is an important test for the atomic structure calculations [12,13] needed to extract weak force parameters from recent atomic parity nonconservation (PNC) experiments in Cs [14]. For a review of the current theoretical methods employed in the calculations see Ref. [15]. The ability to calculate the hyperfine constants implies accurate wave functions at short radius, the same region where the weak interaction occurs. The hyperfine interaction also gives information about nuclear structure; for example, the hyperfine anomaly (Bohr-Weisskopf effect) probes the nuclear magnetization and its variations in a chain of isotopes. It is sensitive to changes in the radial distribution of the neutron magnetization.

In this paper we present our measurements of the $7S_{1/2} \rightarrow 8S_{1/2}$ energy interval and hyperfine splitting of the $8S_{1/2}$ level by resonant two-photon spectroscopy. The dipole-forbidden $7S_{1/2} \rightarrow 8S_{1/2}$ transition is a candidate for a PNC experiment in Fr and the $8S_{1/2}$ level had not been previously located.

To make Fr, a beam from the Stony Brook superconducting linear accelerator impinges on a Au target producing in it $\approx1\times10^7$s $^{210}$Fr, which has a half-life of 3.2 min. We separate the production and trapping regions by extracting the francium atoms out of the gold as ions and transport them about 1 m, where they are neutralized on a heated yttrium surface. The neutral atoms are released into a nonstick dry-film coated glass cell where the magneto-optical trap (MOT) forms. The physical trap consists of a 10-cm-diam Pyrex bulb with six 5-cm-diam windows and two viewing windows 3 cm in diameter. The MOT is formed by six intersecting laser beams each with $7.8$-$\text{mW/cm}^2$ intensity and a magnetic field gradient of $6$ 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, the apparatus is tested extensively with stable Rb. 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 test the excitation and detection schemes needed for the Fr measurements. We have studied the hyperfine splitting of the $6S_{1/2}$ level of $^{85}$Rb to test the method we apply to Fr.

Figure 1 shows the energy levels of the $^{210}$Fr atom relevant for trapping and for the two-photon spectroscopy. A Coherent 899-21 titanium-sapphire laser excites the trapping and cooling transition $7S_{1/2}, F=13/2 \rightarrow 7P_{3/2}, F=15/2$ at $718$ nm. We frequency modulate the trapping laser at $15$ kHz and use lock-in techniques to detect the trap fluorescence. An $817$-nm EOSI 2010 diode laser returns any atoms that leak out of the cooling cycle via the $7S_{1/2}, F=11/2 \rightarrow 7P_{1/2}, F=13/2$ transition.

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**FIG. 1.** Diagram of energy levels of $^{210}$Fr relevant for trapping and two-photon excitation.
We use two different levels, $7P_{3/2}$ or $7P_{1/2}$, for the first step to the $8S_{1/2}$ level (see Fig. 1). Only the $8S_{1/2}$, $F = 13/2$ level can be reached with an electric dipole-allowed transition when the $7P_{3/2}$, $F = 15/2$ level is the intermediate state. An EOSI 2001 diode laser (In, Ga$_1$, As/InP) operating at 1.7 $\mu$m with a linewidth of 110 MHz full width at half maximum (FWHM) excites this transition. Both of the $8S_{1/2}$ hyperfine states can be reached when the $7P_{1/2}$, $F = 13/2$ level is the intermediate state. An EOSI 2010 diode laser operating at 1.3 $\mu$m with a linewidth of 30 MHz FWHM excites this transition. We achieve long-term frequency stability of all the lasers except for the probe by sending part of the beams into a Fabry-Perot cavity along with a frequency-stabilized He-Ne laser. A digital feedback loop locks the lasers to the stabilized He-Ne with less than $0.1\%$ or 20 cm$^{-2}$ accuracy of a few fractions of a percent. A search of the $8S_{1/2}$, $F = 11/2$ state and our quantum-defect fit as a guide. The calculation includes the Fr 10$S_{1/2}$ to 20$S_{1/2}$ levels and ionization potential from ISOLDE [5] and our measurement of the 9$S_{1/2}$ level [9]. The interpolation narrowed the search to a window of less than 1 cm$^{-1}$.

To excite the atoms from the $7P_{3/2}$ level, we split the 1.7-$\mu$m laser beam and send part to the wavemeter and the rest to the trap. The atoms decay from the $8S_{1/2}$, $F = 15/2$ level to the ground state through the $7P_{3/2}$ and $7P_{1/2}$ levels and we detect the fluorescence from the $7P_{1/2}$ level at 817 nm. Since the repumping laser contributes background scattered light at 817 nm, an acousto-optic modulator (AOM) extinguishes the light while a Hamamatsu 636-10 photomultiplier tube (PMT) detects the fluorescence in photon-counting mode [see Fig. 3(a) for the time cycle]. Appropriate interference filters reject light other than at 817 nm. We shape and amplify the pulses and send them to a gated scaler that counts photons when the repumping laser is off. The timing signals come from a digital gate generator (Stanford Research DS400). A computer controls the lasers and reads the wavemeter and the 817-nm scaler. When the 1.7-$\mu$m laser excites the $7P_{3/2}, F = 15/2 \rightarrow 8S_{1/2}, F = 13/2$ transition, the 817-nm scalar counts increase and the 718-nm fluorescence decreases accompanied by a change in the shape of the cloud of atoms.

Figure 2 is a schematic diagram of the apparatus used for the excitation from the $7P_{1/2}$ level. Two lasers excite the atoms to the intermediate state from each of the hyperfine ground states (see Figs. 1 and 2). The 817-nm EOSI 2010 repumping laser excites atoms from the $7S_{1/2}, F = 11/2$ state and a titanium sapphire depumping laser excites atoms from the $7S_{1/2}, F = 13/2$ state. To ensure that both lasers are on resonance AOMs extinguish them simultaneously and a PMT and photon counting electronics detect the fluorescence of the $7P_{1/2}$ level. We adjust the laser frequencies to maximize the 817-nm fluorescence. A second Hamamatsu 636-10 PMT detects 718-nm photons from the decay cascade of the $8S_{1/2}$ level. Two cascaded Gsänger LM0202 electro-optic modulators (EOMs) extinguish the trapping light during the photon counting to reduce the background light [see Fig. 3(b) for the time cycle]. The computer scans the 1.3-$\mu$m laser at a rate of 0.7 MHz/s in the increasing and decreasing frequency directions and records the $8S_{1/2}$ resonance in the 718-nm photon counts. We send part of the 1.3-$\mu$m laser through an EOM that provides 100-MHz sidebands before it enters a 300-MHz free spectral range, temperature-stabilized Fabry-Perot cavity ($=8$ MHz/h drift) that provides a frequency reference. Figure 4 shows resonances of the $8S_{1/2}, F = 13/2$ and $F = 1/2$ hyperfine states together with the etalon signal with the 100-MHz frequency markers.

The probe laser bandwidth determines the resonance widths since the cold atoms have negligible Doppler broadening. The long-term linewidth of the laser is broad. Acoustic noise from the target area couples to the laser producing a linewidth of about 30 MHz over a few seconds. The noise has long-period oscillations that contribute to the broadening and asymmetries observed in Fig. 4. During a scan we monitor the laser with a Fabry-Perot cavity, but we have not been able to obtain a unique deconvolution of the atomic line.

FIG. 2. Diagram of the experimental setup for 1.3-$\mu$m excitation.
shape due to uncertainties in the parameters of the cavity and its alignment. We have performed off-line a series of measurements of the linewidth of the laser using a Fabry-Perot cavity. With the laser subjected to similar noise conditions to those present in the target area, we have found the center of gravity of the cavity resonances to be accurate to better than 0.001 cm$^{-1}$. The difference in the $7S_{1/2} \rightarrow 8S_{1/2}$ transition energy between the two excitation paths is consistent with zero. We add in quadrature the accuracy of the wavemeter ($\pm 0.003$ cm$^{-1}$), the uncertainty for the first step photon ($\pm 0.002$ cm$^{-1}$), and the uncertainty for the second step photon ($\pm 0.002$ cm$^{-1}$) to obtain a total uncertainty of $\pm 0.004$ cm$^{-1}$. The center-of-gravity energy interval between the $7S_{1/2} \rightarrow 8S_{1/2}$ levels of $^{210}$Fr is $19732.523 \pm 0.004$ cm$^{-1}$.

The ab initio calculations use many-body perturbation theory to calculate the ionization energy of the $8S_{1/2}$ level. We use the ionization energy of the $7S_{1/2}$ level from Ref. [5] to compare our result with the calculations of Dzuba et al. (19739 cm$^{-1}$) [1] and Johnson (19665 cm$^{-1}$) [2]. Their predictions agree with our measurement to better than 4 parts in $10^3$.

The $8S_{1/2}$ level completes the lower part of $nS_{1/2}$ series of francium. The energies of the $nS_{1/2}$ levels are well represented by the Rydberg series

$$E_{nS} = E_{\text{ion}} - \frac{R_m}{n_{\text{eff}}}^2,$$

where $E_{\text{ion}}$ is the ionization energy of the ground state, $E_{nS}$ is the energy of the $nS_{1/2}$ level measured from the ground state, and $R_m$ is the reduced mass Rydberg constant. Equation (1) assigns an effective quantum number $n_{\text{eff}}$ to the known francium $nS_{1/2}$ levels. We use an iterative algorithm to fit the quantum defect parameters $\delta_0$, $\delta_2$, $\delta_4$, and $\delta_6$ to the measured energies using

$$n_{\text{eff}} = n - \delta = n - \left( \delta_0 + \frac{\delta_2}{(n - \delta)^2} + \frac{\delta_4}{(n - \delta)^4} + \frac{\delta_6}{(n - \delta)^6} + \cdots \right),$$

where $\delta$ is the quantum defect. Table I gives the quantum-defect parameters from three fits: rows 1 and 2 by Arnold et al. [5] and 3 from this work. Row 1 includes the $7S_{1/2}$ and $10S_{1/2} \rightarrow 22S_{1/2}$ levels. Row 2 includes the $10S_{1/2} \rightarrow 22S_{1/2}$ levels. Row 3 includes the $7S_{1/2} \rightarrow 22S_{1/2}$ levels. Figure 5 compares the measured energy levels of the $nS_{1/2}$ series to our quantum-defect calculation. We correct for the isotope and hyperfine shifts from $^{210}$Fr to $^{212}$Fr to use the measurements of Arnold et al. [5] in our fit.

We obtain the $8S_{1/2}, F = 11/2$ to $F = 13/2$ hyperfine splitting from the wavemeter readings of the resonances with the $7P_{1/2}$ as the intermediate level. We use the center of gravity of the line from the wavemeter readings and assign an un-
which, according to the authors, have an accuracy not worse than 3\%, and Owusu et al. [4]. We use the measurements of the magnetic moments of Ref. [21] and the nuclear spins to obtain a hyperfine constant $A$ for $^{210}\text{Fr}$ from the theoretical calculations. The hyperfine separation of the $8S_{1/2}$ level of $^{210}\text{Fr}$ is $10 256\pm 7$ MHz, which gives a hyperfine constant $A$ of $1577.8\pm 1.1$ MHz with an overall fractional accuracy in the $A$ hyperfine constant of $7\times 10^{-4}$.

We have performed two-photon spectroscopy of francium in a MOT using the $7p$ levels as intermediate resonant steps to measure the energy interval between the $7S_{1/2}$ and $8S_{1/2}$ levels. The location of the $8S_{1/2}$ level is necessary for the continuing effort to study parity nonconservation in francium. We have also measured the hyperfine splitting of the $8S_{1/2}$ level of $^{210}\text{Fr}$ and compared the result with ab initio calculations. Measurements of the same hyperfine splitting in other isotopes could reveal nuclear structure information that can be sensitive to changes in the neutron distribution.

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