Measurement of the $7p\,^2P_{3/2}$ Level Lifetime in Atomic Francium

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We present the first measurement of an atomic radiative lifetime in Fr. We use a time-correlated single photon counting technique with a cold sample of ^{210}Fr atoms in a magneto-optic trap. The results are a precision experimental test of the atomic many-body perturbation theory applied to the heaviest alkali. The lifetime for the $7p\,^2P_{3/2}$ level of 21.02(16) ns gives a value for the reduced transition matrix element between the levels $7s\,^2S_{1/2} \rightarrow 7p\,^2P_{3/2}$ of $5.898(22)a_\infty$ atomic units. [S0031-9007(97)03279-1]

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Francium remains the least studied of alkali atoms, but is of great interest as the heaviest "simple" atom. Some of the energy levels and hyperfine splittings have been measured [1–3] and compared to *ab initio* calculations [4–6]. The difficulty in making measurements resides in the scarcity of this unstable element that requires the use of accelerators or radioactive sources for production. Until now none of the radiative lifetimes have been measured. The lifetimes are a sensitive test of the atomic wave functions since they give the transition matrix elements between energy levels. Fr is an ideal atom to study with the recently developed techniques of many-body perturbation theory (MBPT). *Ab initio* calculations give the location of energy levels, lifetimes, and hyperfine structure constants. Each of these is sensitive to a different property of the calculated wave function. Fr has a structure similar to Cs but has 32 more electrons in a closed radonlike shell, making it a more difficult atom to calculate. Relativistic corrections are very important.

The present atomic parity nonconservation (PNC) efforts in cesium and in thallium are reaching experimental uncertainties of fractions of a percent [7,8]. High precision calculations are necessary for the extraction of the weak charge from atomic PNC measurements [6,9]. Fr is an attractive candidate for a future precision test of the standard model at low energy, because the PNC effect is 18 times larger in Fr than in Cs [6]. Since the methods used for calculating Cs and Fr are the same, measurements of the atomic properties of Fr, and comparisons with theory, are also a very sensitive test for the accuracy of the calculations in Cs [10].

In this Letter we present the measurement of the radiative lifetime of the $7p\,^2P_{3/2} \rightarrow 7s\,^2S_{1/2}$ (D_2 line) and compare it with state of the art *ab initio* calculations for the transition matrix elements. The measurement tests the calculated wave functions at large radius. It is complementary to studies of the hyperfine constants that are most sensitive to the wave functions at the nucleus [1,5]. We recently located the $9S_{1/2}$ level [3] and compared the measured energy to *ab initio* predictions [6] with an agreement within the theoretical accuracy of 10^{-3}. Our present lifetime measurement opens the experimental exploration of the Fr atom beyond the location of its energy levels.

The MBPT calculations for the Fr matrix elements [6,11] start with a relativistic Hartree-Fock method. They use different techniques to improve the calculation, taking into account higher-order corrections. Some of the higher-order corrections are added empirically based on what is known in Cs. The values obtained and the accuracies claimed provide a set of benchmark values for comparison with experiments and with future "all-order" *ab initio* calculations.

Recent approaches to precise lifetime measurements in alkali atoms include highly refined fast beam experiments [12,13], direct linewidth measurement with cold-trapped atoms [14], and time-correlated single photon counting [15]. A novel approach in the field has been the spectroscopy of a molecule composed of two identical atoms, one in the ground state and the other in a resonant excited state. The dipole matrix element that enters in the lifetime dominates the behavior of the interatomic potential at long distances and can be determined from an analysis of the vibrational spectrum of molecules formed in very cold samples of atoms [16–18].

We employ a technique to measure the lifetime that efficiently uses the limited number of atoms available. We capture Fr atoms in a magneto-optic trap (MOT) on-line with the superconducting LINAC at Stony Brook. The trap has been developed for working with radioactive alkalis. The specific details on the production and trapping of ^{210}Fr can be found in Refs. [19,20]. We use time-correlated single photon counting techniques in a MOT with cold atoms. The trapping laser is quickly turned off, and we observe the free exponential decay of the fluorescence from the excited atoms in the trap. The cold atoms move less than 0.1 μm during the decay.

Figure 1 shows the atomic energy levels of the ^{210}Fr atom relevant for the measurement. The glass-cell MOT operates in the cycling transition $7s\,^2S_{1/2}$, $F = 13/2 \rightarrow 7p\,^2P_{3/2}$, $F = 15/2$ at 718 nm. A laser tuned to the $F = 11/2$ ground state to $F = 13/2$ excited state repumps the atoms. The MOT is formed by six intersecting laser

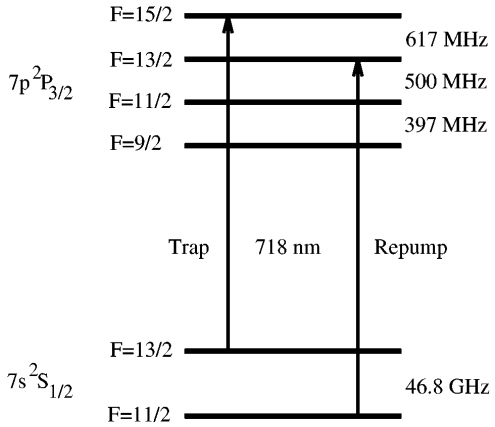

FIG. 1. Energy levels of ^{210}Fr relevant for trapping and for the lifetime measurement.

beams each with $1/e^2$ (power) diameter of 4 cm and power of 150 mW, with a magnetic field gradient of 9 G/cm. The trap has a lifetime of approximately 20 s.

Since Fr is unstable, a Burleigh WA-1500 wavemeter and an I_2 cell provide the frequency references for the transitions. The trapping laser is frequency modulated with an amplitude of 4 MHz at a frequency of 14.5 kHz. The repumper laser is modulated in frequency with an amplitude of 500 MHz and a frequency of 4 kHz. Lock-in detection allows the rejection of the laser light scattered from the glass cell while measuring the fluorescence from the captured atoms. A $f/2$ optical system collects the fluorescence of the trapped atoms onto a Hamamatsu R636-10 photomultiplier tube (PMT), and the signal is demodulated by a Stanford Research SRS 852 lock-in detector. This signal permits the continuous monitoring of the trap during the lifetime measurements.

The main trapping laser beam passes through two electro-optic modulators (EOM), Gsänger LM0202, before it enters the trap. The light is turned on and off with a Gaussian $1/e$ (power) fall time of 9 ns and extinction ratio better than 800 : 1 (after 30 ns). The repumper laser is turned off synchronously using an acousto-optic modulator (AOM). The timing sequence for the detection comes from a Stanford Research S400 digital gate generator (DGG). A second Hamamatsu R636-10 PMT with similar $f/2$ collection optics views the trap and operates in a photon-counting mode for the lifetime measurements. An interference filter and a small aperture minimize scattered background light into the PMT. This is necessary to ensure that the PMT is not saturated when the trapping laser is on. The photoelectric pulse, after shaping, provides the start pulse to the time-to-amplitude converter (TAC) Ortec 467, and a gate signal from the DGG, synchronized with the EOM, provides the stop pulse. This time-reversed mode reduces dead time in the electronics. The output of the TAC goes to a multichannel analyzer (MCA) Canberra 35 Plus where the data is accumulated, binned, and stored for later processing (see Fig. 2).

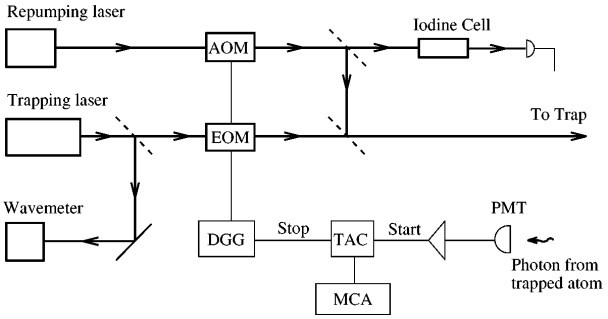

FIG. 2. Block diagram of the apparatus.

Each data collection run consists of two parts. With Fr atoms in the trap, we collect a histogram of the fluorescence for thirty minutes by turning off the light for 500 ns at a rate of 100 kHz. Then we collect a background histogram for the same period of time except that we shift the repumper frequency away from resonance so that no atoms accumulate in the trap. In this way we have the same amount of scattered light. We maintain the same level of accelerator beam current while taking a background run, in case there are counts due to the radioactivity present in the nuclear reaction. Each pair of data samples is analyzed together (see Fig. 3 for an example of the raw data). We first apply an analytical correction to all data points (sometimes called pulse pileup) arising from the fact that the early events are preferentially counted [21]. The correction modifies the extracted lifetime of Fr by $+0.05\%$. The time calibration of the TAC and MCA has an uncertainty of $\pm 0.04\%$. Their amplitude and time linearity, and the frequency jitter of the cycle contribute at most $\pm 0.1\%$ (see Table I).

To obtain a data sample (trace *a* in Fig. 3), we take the signal (trace *b* in Fig. 3) and subtract the background (trace *c* in Fig. 3). We then fit the data for a given number of channels, starting at a time where the light is off, to a decaying exponential with a constant background. At the bottom of Fig. 3 are the residuals of such a fit divided by the statistical uncertainty of each point. From all the runs we obtain a Fr lifetime with a standard deviation of their mean of $\pm 0.24\%$.

We also fitted the data to the convolution of the well-characterized instrument function with a single exponential. The convolution allows us to use points closer to the turn-off time and gives the same value for the lifetime within the statistical uncertainty as that obtained with a single exponential. The value of the constant background from the fits is very small (less than 5 counts), consistent with zero. Although the quality of each individual fit is very good (reduced $\chi^2 \approx 1$), the particular value obtained for the lifetime depends on the number of channels fitted. For each individual Fr run, we change the number of fitted channels both at the begining and at the end of the data sample. We vary the starting point of the fit with respect to the light turn-off time by 0.8τ and the end point by as many as 5τ. We assign an uncertainty of $\pm 0.6\%$,

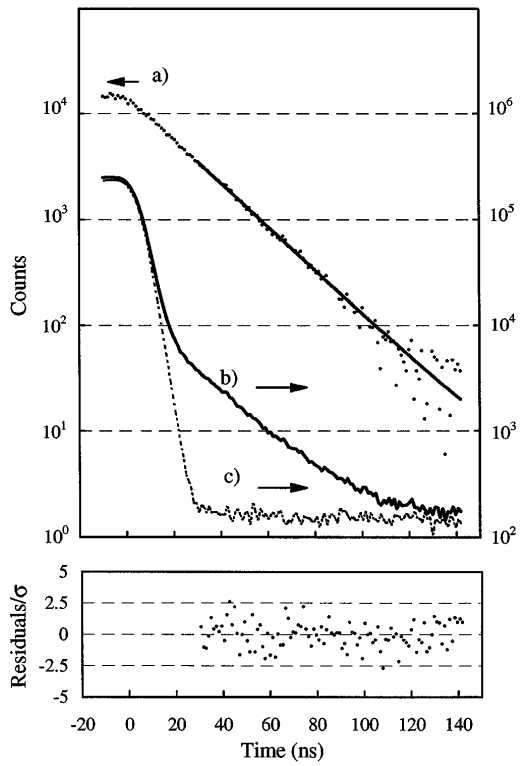

FIG. 3. Decay curves of Fr $7p\,^2P_{3/2}$ level. Trace a is the subtraction of the direct measurement with Fr in the trap (trace b) and the background (trace c). The straight line is a pure exponential fit to trace a. The bottom shows the residuals of the fit divided by the statistical uncertainty of each point. The reduced $\chi^2 = 1.01$.

which accounts for the variation in the lifetime from the truncation in the fit [15].

The number of atoms in the trap (well depth $\approx$0.5 K) is small ($N \approx 1000$), and the typical diameter of the trap is $\leq$1 mm. This minimizes the possibility of multiple absorption. We perform the measurement with different values of the trapping magnetic field gradient and see no change in the measured lifetime.

We have considered the possibility of hyperfine and Zeeman quantum beats, but we have not observed any in the fluorescence signal. The hyperfine splitting between the $F = 15/2$ and $F = 13/2$ is 617 MHz. The laser linewidth ($\leq$500 kHz) is too narrow and the pulse length (10 μs) and fall time (9 ns) too long to excite coherently both states. Zeeman quantum beats are a possible source of systematic shifts. The position dependent force in a MOT requires a gradient of the magnetic field in three dimensions. For our trap an atom that fluoresces may be in a magnetic field of at most 0.5 G. The inhomogeneous magnetic field gives each atom a different Larmor frequency, and the different polarizations available from all the laser beams will tend to average out this effect when we count photons without a polarizer in front of the detector. Nevertheless, we have calculated the contribution of a Zeeman quantum beat to the lifetime of ^{210}Fr for a geometry that allows coherences between excited state m_F sublevels differing by two [22] in an average field of 0.25 G (beat frequency 187 kHz). In our calculation the fitted lifetime is 0.03% shorter than that from the decay free from quantum beats. We take this number as a systematic error. In order to confirm the absence of other oscillations or correlations in the data, we Fourier transform the residuals of the fit and look for any possible remanent frequency. The spectrum shows only noise with no distinguishable peaks.

TABLE I. Error budget for the lifetime of the D_2 lines of Rb and Fr in percentage.

Error	Rb (%)	Fr (%)
Systematic		
TAC-MCA nonlinearity	$\pm$0.10	$\pm$0.10
Time calibration	$\pm$0.04	$\pm$0.04
Truncation error	$\pm$0.24	$\pm$0.60
Zeeman quantum beat	$\pm$0.16	$\pm$0.03
Other	$\pm$0.23	$\pm$0.39
Total systematic	$\pm$0.37	$\pm$0.72
Statistical	$\pm$0.07	$\pm$0.24
Sum in quadrature	$\pm$0.39	$\pm$0.76

To search for other possible shifts in the lifetime, we have measured the lifetime of the $5p\,^2P_{3/2}$ level of ^{87}Rb using exactly the same apparatus and technique. A Rb dispenser sprays atoms onto the surface of the target, and the Rb follows the same path as Fr in our system [20]. With Rb in the trap, we have varied the number of atoms by decreasing the repumper power. We changed the modulation amplitude of the trapping laser. We moved the position ($\pm$ one trap diameter) of the trap by imbalancing the light in one arm of the trap, while keeping the detector fixed, and we changed the magnetic field gradient. For all these tests we found no appreciable effect within the statistical error of $\pm$0.23% of the lifetime from a single data run. We take this number as a limit on other systematic errors. The standard deviation of the mean lifetime of Rb is $\pm$0.07%. The truncation error of the fit is $\pm$0.24%. The effect on the lifetime of the possible quantum beats (frequency of 467 kHz) calculated for ^{87}Rb is 0.16%, and we add it as a systematic error. The Fourier transform of the residuals shows no distinguishable peaks.

The combination of the errors in quadrature gives an uncertainty in the Rb measurement of $\pm$0.39%. We add this same error to the Fr value as a limit for other systematic contributions.

The result of our lifetime measurement of the $7p\,^2P_{3/2}$ level of Fr is 21.02(16) ns (1σ) and the uncertainty (see Table I) is a combination in quadrature of the contributions described above. Our measurement on the $5p\,^2P_{3/2}$ level of Rb used to check the method is 26.20(10) ns in very good agreement with the recent most precise measurement of Voltz and Schmoranzer that gives a value of 26.24(4) ns [12].

From our measurement of the atomic lifetime in Fr, we can determine the matrix elements. The excited $7p\,^2P_{3/2}$

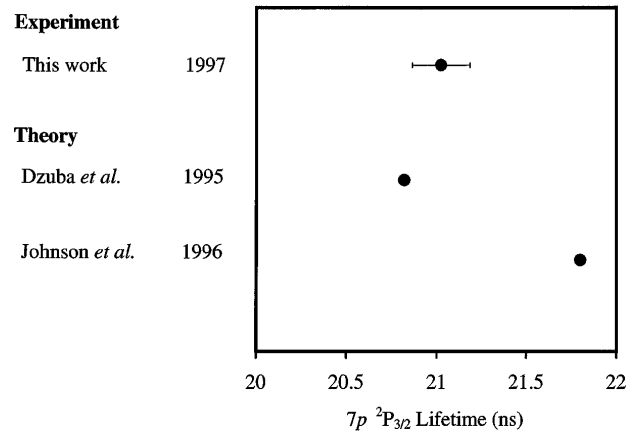

FIG. 4. Comparison of the $7p\,^2P_{3/2} \rightarrow 7s\,^2S_{1/2}$ transition lifetime measurement with *ab initio* calculations from Refs. [6,11].

level has a single decay mode, and its lifetime is related to a single reduced radial matrix element between the excited and ground states by

$$\frac{1}{\tau_{J'\rightarrow J}} = \frac{4}{3}\,\frac{\omega^3}{c^2}\,\alpha\,\frac{|\langle J\|r\|J'\rangle|^2}{2J'+1}\,,$$

where ω in the transition energy is divided by $\hbar$, c is the speed of light, α is the fine-structure constant, J' and J are, respectively, the excited and ground state angular momenta, τ is the excited state lifetime, and $|\langle J\|r\|J'\rangle|$ is the reduced matrix element. Using the experimentally measured transition energy [20], the 21.02(16) ns lifetime gives $5.898(22)a_\infty$ in atomic units for the $7s\,^2S_{1/2} \rightarrow 7p\,^2P_{3/2}$ reduced matrix element, with a_∞ the Bohr radius.

Figure 4 presents the graphical comparison between this measurement and the MBPT *ab initio* calculations [6,11]. The theoretical lifetime values use the calculated matrix element and the measured transition energy. The accuracies assigned to the predictions are of the order of a percent and overlap the measured value. This result gives increasing confidence to the state of the art atomic structure calculations.

We have measured the radiative lifetime of the Fr D_2 line in a MOT. The precision is enough to determine the transition matrix element to 0.37%. This matrix element is a new stringent test of the relativistic many-body calculations in alkali atoms. This is a new piece of experimental data that will hopefully motivate even higher-order *ab initio* calculations. This measurement tests the calculation techniques used for the interpretation of the Cs PNC experiments for a more difficult atom. The result enhances confidence in the current calculations and increases the need for more accurate calculations in the future.

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