Lifetime measurement of the 9*s* level of atomic francium

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We use two-photon resonant excitation and time-correlated single-photon counting techniques on a sample of ^{210}Fr atoms confined and cooled in a magneto-optical trap to measure the lifetime of the 9*s* excited level. Direct measurement of the decay through the $7P_{3/2}$ level at 851 nm yields a lifetime of 107.53 ± 0.80 ns.

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Interest in the spectroscopic properties of the *s* excited states of Fr is intimately related to the atom's potential to perform fundamental studies of discrete symmetries.[1] The pioneering work on Fr at the On-Line Isotope Mass Separator[2] (ISOLDE) opened the possibility of cooling and trapping[3,4] this radioactive heavy alkali in sufficient quantities to go beyond energy levels and hyperfine splittings in its atomic structure studies.[5] Understanding of its *p* and *d* states is beginning to reach a level comparable with that of the other alkalis. Of particular interest is the quantitative knowledge reached by the atomic theory calculations of matrix elements[6–8] and its agreement with our measurements, which strengthen the possibility of a parity nonconservation (PNC) experiment in a chain of Fr isotopes.

This Letter reports our measurement of the lifetime of the 9*s* level, to our knowledge the only lifetime of an *s* excited level in Fr ever measured, which presents a new challenge to the most sophisticated techniques of *ab initio* calculations with the many-body perturbation theory. The electric dipole forbidden transitions between *s* states in alkali metals are ideal for PNC measurements, as shown by the work of Wood *et al.*[9] with Cs. Fr is yet to be used in PNC measurements, but understanding its excited-state properties is one way to test the accuracy of the atomic theory calculations that are necessary for extracting information about the weak force from future measurements of Fr.[6,8]

The production, cooling, and trapping of Fr on line with the superconducting linac at Stony Brook has been described previously.[1,10] Briefly, a 100-MeV beam of ^{18}O ions from the accelerator impinges on a Au target to make ^{210}Fr. We extract $\sim 1 \times 10^6$ Fr ions/s from the Au and transport them ~15 m to a cold Y neutralizer, where we accumulate the Fr atoms. We then close the trap with the neutralizer and heat it for 1 s (~1000 K) to release the atoms into the dry-film-coated glass cell, where they are cooled and trapped in a magneto-optical trap (MOT) with 10^4–10^5 atoms captured on average.

Figure 1 shows the energy levels of ^{210}Fr that are relevant for trapping and lifetime measurements. A Coherent 899-21 Ti:sapphire laser operating at 718 nm excites the trapping and cooling transition ($7S_{1/2}, F = 13/2 \rightarrow 7P_{3/2}, F = 15/2$). A Coherent 899-01 Ti:sapphire laser operating at 817 nm repumps any atoms that leak out of the cooling cycle via the $7S_{1/2}, F = 11/2 \rightarrow 7P_{1/2}, F = 11/2$ transition.

We measure the lifetime of the 9*s* level with time-correlated single-photon counting.[11] A short pulse of resonant laser light populates an excited state at $t = 0$. The histogram of arrival times, with respect to the excitation pulse of the spontaneously emitted photons directly shows the exponential decay of the state. This method has been used, for example, to measure lifetimes of atoms in beams,[12] atoms in vapor cells,[13] trapped single ions,[14] and atoms in a MOT.[5]

Figure 2 shows the basic elements of the apparatus and timing. All the lasers are turned on and off at the appropriate times to optimize the measurement of the excited-state lifetime. The pulsed probe excitation (100–400 ns long) from the $7P_{1/2}, F = 11/2$ level to the $9S_{1/2}, F = 13/2$ level comes from another Coherent 899-21 Ti:sapphire laser operating at 744 nm that is amplitude modulated with an extinction ratio of better than 400:1 (decay time less than 10 ns) by cascaded electro-optical and acousto-optical modulators (AOMs). To maximize the signal, we move the population to the $7S_{1/2}, F = 11/2$ level with a depumping laser ($7S_{1/2}, F = 13/2 \rightarrow 7P_{3/2}, F = 13/2$), then the repumper provides the first step (through the $7P_{1/2}, F = 11/2$ level), and the probe provides the second step.

The Burleigh WA-1500 wavemeter monitors the wavelength of the lasers to approximately ± 0.001 cm^{-1} while a computer-controlled scanning Fabry–Perot cavity monitors and holds the long-term frequencies of all the lasers at the megahertz level.[5] A 1:1 imaging system ($f/3.9$) collects the wavelength-selected fluorescence on both a high-speed cooled CCD camera (Roper Scientific, MicroMax 1300YHS-DIF) for the trapping fluorescence at 718 nm (Andover 720FS10-50) and a photomultiplier tube (Hamamatsu R636) for the lifetime measurement at 851 nm (Andover 850FS10-50). The signal from the CCD camera gives us the number of trapped atoms. We amplify and discriminate the pulses from the photomultiplier tube before sending them to the pulse-processing electronics. We keep the rate of photon counts low to prevent double-pulse events and reduce dead-time systematic effects in the electronics.

The measurement operates on the 10-μs cycle sketched at the bottom of Fig. 2. We excite a small fraction of the atoms in the $7P_{1/2}$ level with the 744-nm

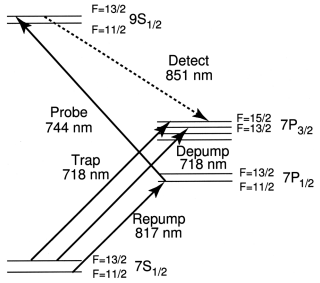

Fig. 1. ^{210}Fr energy levels and lasers relevant to the trapping operation and lifetime measurements.

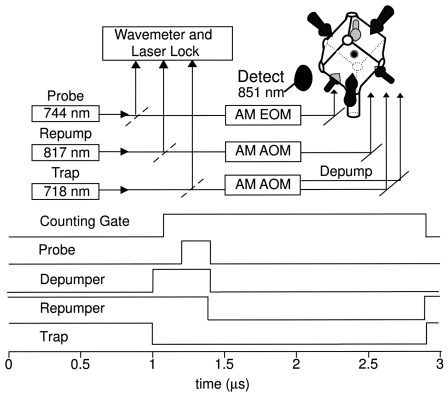

Fig. 2. Block diagram and timing sequence of the experiment. AM, amplitude modulated; EOM, electro-optic modulator.

probe laser. After the probe pulse, we detect the decay of the fluorescence through the $9S_{1/2} \rightarrow 7P_{3/2}$ decay channel at 851 nm for 1000 ns. We also monitored the indirect decay through the $8p \rightarrow 7s$ transitions at 423 and 433 nm.[15]

A Berkeley Nucleonics BNC 8010 pulse generator provides the timing sequence for the chopping and detection. Detected fluorescence photon pulses start an Ortec 467 time-to-amplitude converter (TAC), the TAC stop pulse comes from the master pulse generator. Starting the TAC with a fluorescence photon eliminates the accumulation of counts from cycles with no detected photons. A multichannel analyzer (MCA) bins the TAC output to produce a histogram of the events.

We accumulate data for ~1000 s with atoms in the trap and the probe laser on resonance. See Fig. 3 for an example of the data. We then repeat the counting measurement without atoms to obtain a background from the long imperfect turn-off of the trap laser that is focused to a line at the AOM position instead of to a point to avoid intensity damage to the AOM. The trapping beams are the strongest, and despite the interference filters used, there is some leakage into the phototube. This gives us a linear background with a slope of 4 counts every 1000 s for 1000 channels. We include it in the fits of the data to obtain good reduced χ^2_ν (~1 ± 0.05).

The time calibration of the TAC and MCA has an uncertainty of ±0.005%. The time scale is linear to ±0.04% over the entire range of channels with data. We measure the differential nonlinearity of the TAC and MCA and find that the binned counts are consistent with a flat line within 1×10^{-6} per channel and do not influence the results.

For a given cycle the TAC can register only one photon. A correction to the raw data accounts for the preferential counting of early events.[11] Low count rates, as in our case, keep this correction small. The correction alters the fitted lifetime of a typical run by less than 0.1%.

We do not find a statistically significant variation in the fitted lifetime depending on the beginning and ending points used to fit the data. The variation is always smaller than what is statistically expected, so we do not report a truncation error. We check this with Monte Carlo simulations and find that they are consistent with our measurement.

We search for quantum beats in the fluorescence decay signal but do not observe any. We measure the lifetime as a function of the magnetic gradient and extrapolate to a zero field. The extrapolation is consistent with no change. We calculate an upper bound for the expected quantum beat signal following Ref. 5 and put an upper limit of ±0.2% in the error budget.

The density of the atoms in the trap is low (~10^9/cm^3). In this regime, effects such as radiation trapping, superfluorescence, and quenching due to collisions do not alter the measured lifetime. We carry out measurements of the $7s$ level of Rb in the same system and with the same apparatus to study the other sources of error and find that they are consistent with our previous studies of the lifetimes of the $7p$ and $7d$ levels in Fr and the $5p$ levels in Rb.[5] An imbalance of the trap laser beams that displaces the trap center by one trap diameter (~0.2 mm) does not significantly affect the measured lifetime through Zeeman quantum beats or any other means beyond the quoted uncertainties.

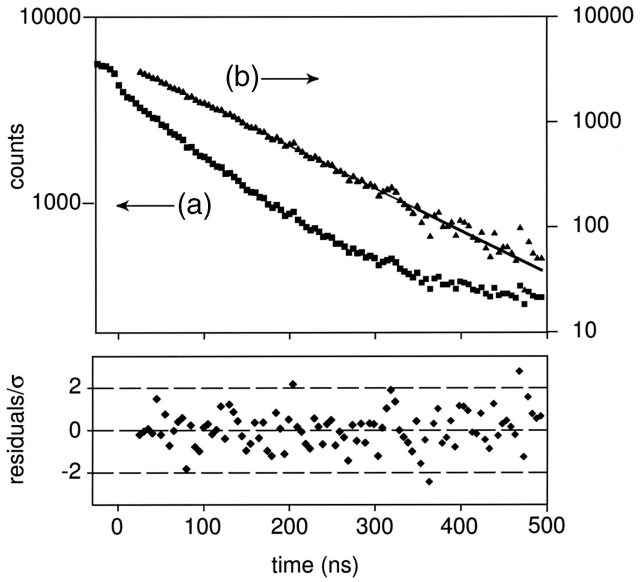

Fig. 3. Decay curve of the $9S_{1/2}$, $F = 13/2$ level with fit residuals. (a) Binned data. (b) Counts minus the background. The line is a fit to an exponential decay. The lower plot shows the residuals of the fit divided by the statistical uncertainty of each point.

Table 1. Error Budget for 9*s* Lifetime Measurement

Error	%
Statistical	±0.72
Time calibration	±0.01
TAC/MCA nonlinearity	±0.04
Quantum beats	<±0.2
Total	±0.75

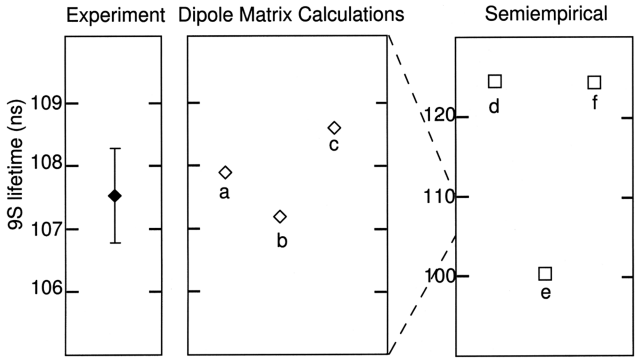

Fig. 4. Comparison of the measured lifetime with theoretical predictions from radial matrix elements a,[7] b,[16] c,[17] and semiempirical calculations d,[19] e,[20] f.[21]

Table 1 contains the error budget for the lifetime measurements. Combining the uncertainties in quadrature yields a total uncertainty of ±0.75%. This yields a lifetime result of 107.53 ± 0.80 ns for the 9*s* level of Fr.

Figure 4 compares our lifetime measurement results with theoretical predictions. The $9S_{1/2}$ level can decay to the $7P_J$ and $8P_J$ ($J = 1/2, 3/2$) levels. We use the appropriate reduced radial matrix elements from the *ab initio* many-body perturbation theory[7,16] or a one-active electron potential[17] with our measured energies[18] to carry out the calculation of the lifetime.[5] The figure also shows semiempirical calculations of the lifetime.[19–22]

The calculations from the radial matrix elements make predictions within our experimental uncertainty. The *ab initio* calculations of Safronova *et al.*[7] and Dzuba and Flambaum[16] are impressive given the complexity of the atom where large correlation contributions add with opposite sign. The prediction of Marinescu *et al.*[17] is also very good and serves as a quantitative test for their work on van der Waals coefficients for Fr.

We have used two-photon excitation and time-correlated single-photon counting techniques on a sample of cold ^{210}Fr atoms confined in a MOT to measure the lifetime of the $9S_{1/2}$ excited level with a 0.75% error. This lifetime is a quantitative test of calculations of radial matrix elements for an excited *s* level in Fr and shows excellent agreement.

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