Lifetime Measurement of the 8s Level in Francium

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(Dated: January 5, 2018)

We measure the lifetime of the 8s level on a magneto-optically trapped sample of $^{210}\text{Fr}$ atoms with
time-correlated single-photon counting. The $7P_{1/2}$ state serves as the resonant intermediate level
for two-photon excitation of the 8s level completed with a 1.3 $\mu$m laser. Analysis of the fluorescence
decay through the the $7P_{1/2}$ level gives $53.30 \pm 0.44$ ns for the 8s level lifetime.

PACS numbers: 32.70.Cs, 32.80.Pj, 32.10.Dk

We present in this letter a measurement of the 8s level lifetime of francium; the heaviest and most relativistic
of alkali atoms. It is a test of the modern techniques of ab initio calculations using many-body perturbation
theory (MBPT) $^1, 2$. Fr is yet to be used in Parity Non-conservation (PNC) measurements $^3$, but work towards
that goal requires the understanding of the excited state properties of the atom. It is important to ensure
that our quantitative understanding of the atomic structure of Fr is as good as that of lighter alkali e.g. Cs
where PNC experiments have achieved resolution to extract weak force parameters $^4$. Quantitative measurements
on Fr and comparison with theoretical calculations validate the same MBPT techniques used for Cs
and other atoms with a more relativistic atom where correlations from the 87$^e$ electrons are large. The 8s state is
the preferred candidate for an optical PNC measurement; the dipole forbidden excitation between the $7S_{1/2}$ ground
state and the first excited $8S_{1/2}$ state, becomes allowed through the weak interaction.

The lifetime $\tau$ of an excited state is determined by its individual decay rates, $1/\tau_i$, through the matrix element
associated with the $i$ partial decay rate. The connections between lifetime, partial decay rates and matrix elements are:

$$\frac{1}{\tau} = \sum_i \frac{1}{\tau_i} \frac{\langle |J||r||J'\rangle^2}{2J' + 1} \; \cdot \; \frac{\hbar}{\omega} \; ,$$

where $\omega$ is the transition energy divided by $\hbar$, $c$ is the speed of light, $\alpha$ is the fine-structure constant, $J'$ and $J$
are respectively, the initial and final state angular momenta, and $\langle |J||r||J'\rangle$ is the reduced matrix element $^5$. Equation
$^6$ links the lifetime of an excited state to the electronic wavefunctions of the atom. The comparison of measurements
with theoretical predictions test the quality
of the computed wavefunctions specially at large distances from the nucleus due to the presence of the radial operator.

The lifetimes of the low lying states in Fr are reaching a level of precision comparable to that of the other alkalis
$^6, 7, 8$. The atomic theory calculations for these transitions $^8, 9, 10$ predict the lifetimemes measured with
impressive agreement, strengthening the possibility of a PNC experiment in a chain of francium isotopes.

We use the method of time correlated single photon counting to obtain the lifetime of the 8s level in Fr in a
magneto-optical trap (MOT). We populate the 8s level with a two photon transition, then we turn off the excitation
suddenly and observe the exponential decay through the fluorescence photons $^11$.

The production, cooling and trapping of Fr on-line with the superconducting linear accelerator at Stony
Brook has been described previously $^12$. Briefly, a 100 MeV beam of $^{18}$O ions from the accelerator impinges on
a gold target to make $^{210}$Fr (radioactive half life 3 min). We extract $\sim 1 \times 10^6$ francium ions/s out of the gold
and transport them 15 m to a cold yttrium neutralizer where we accumulate the Fr atoms. We then close the trap with
the neutralizer and heat it for one second ($\sim 1000$ K) to release the atoms into the dry-film coated glass cell where
they are cooled and trapped in a MOT. The cycle of accumulating and trapping repeats every 20 s.

Figure $^13$ shows the states of $^{210}$Fr relevant for trapping and lifetime measurements. A Coherent 899-21 titanium-
sapphire (Ti:Sapph) 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 899-21 Ti:Sapph laser operating at 817 nm repumps any atoms that leak out of
the cooling cycle via the $7S_{1/2}, F = 11/2 \rightarrow 7P_{1/2}, F = 13/2$ transition. The first photon for the $7S_{1/2} \rightarrow 8S_{1/2}$
transition comes from a Coherent 899-01 Ti:Sapph at 817 nm, it populates the $7P_{3/2}, F = 13/2$ state. The second
photon at 1.3 $\mu$m originates from an EOSI 2010 diode laser to excite the $7P_{3/2} \rightarrow 8S_{1/2}$ transition.

A Burleigh WA-1500 wavemeter monitors the wavelength of all lasers to about $\pm 0.001$ cm$^{-1}$. We lock the
trap, first photon and repumper lasers with a transfer lock $^13$, while we lock the second photon laser with the aid of a Michelson interferometer that is itself locked to the frequency stabilized HeNe laser used in the transfer
lock.

The MOT consists of three pairs of retro-reflected beams, each with 15 mW/cm$^2$ intensity, 3 cm diameter
(1/e intensity) and red detuned 31 MHz from the atomic
resonance. A pair of coils generates a magnetic field gradient of 9 G/cm. We work with traps of \( \approx 10^4 \) atoms, a temperature lower than \( 300 \) \( \mu \)K, with a diameter of 0.5 mm and a typical lifetime between 5 and 10 s.

Figure 2 displays the timing sequence for the excitation and decay cycle for the measurement. Both lasers of the two photon excitation are on for 50 ns before they are switched off, while the counting electronics are sensitive for 500 ns to record the excitation and decay signal. The trap laser turns off 500 ns after the two-photon excitation. We repeat the cycle at 100 KHz.

We turn the trap light on and off with an electro optic modulator (EOM) (Gässinger LM0202) and an acousto optic modulator (AOM) (Crystal Technology 3200-144). The combination of the two gives an extinction ratio of better than 1600:1 after 500 ns. AOMs modulate the repumper and the first photon (817 nm) light (Crystal Technology 3200), they have extinction ratios of 109:1 and 26:1 30 ns after the pulse turns off respectively. We couple the 1.3 \( \mu \)m laser into a single mode optical fiber pass it through a 10 Gbits/s lithium niobate electro-optic fiber modulator (Lucent Technologies 2623N), then amplify it (Iphenix IPSAD1301) and again modulate it with a second electro-optic fiber modulator (Lucent Technologies 2623N); the result is an on-off ratio of better than 1000:1 in a time of 20 ns.

A 1:1 imaging system (f/3.9) collects the fluorescence photons onto a charge coupled device (CCD) camera (Roper Scientific, MicroMax 1300VHS-DIF). We monitor the trap with the use of an interference filter at 718 nm in front of the camera. A beam-splitter in the imaging system sends 50% of the light onto a photo multiplier tube (PMT) (Hamamatsu R636). An interference filter at 718 nm in front of the PMT reduces the background light other than fluorescence from the cascade through the 7\( P_{1/2} \) level decay back to the ground state 7\( S_{1/2} \).

After we turn off the excitation lasers, the atom returns back to the ground level using two different decay channels (see Fig. 1): First, by emitting a 1.3 \( \mu \)m photon it decays back to the 7\( P_{1/2} \) state and fluoresces 817 nm light to return to the 7s ground level. The second possible decay channel is the 8s \( \rightarrow \) 7\( P_{3/2} \) transition followed by the decay to the 7s ground level. The 1.7 \( \mu \)m fluorescence from the first step of this decay is unobserved, but we detect 718 nm light from the second part of the decay. With the known lifetime of the 7\( P_{3/2} \) state, it is possible to extract the 8s level lifetime from the cascade fluorescence decay.

We amplify (Ortec AN106/N) the current pulses from the photon detections in the PMT. We gate (EG&G LG101/N) and send them to a constant fraction discriminator (Ortec 934). The output starts a gated time-to-amplitude converter (TAC) (Ortec 467) that we stop with a fixed time delay pulse after the two-photon excitation. We use a multichannel analyzer (MCA) (EG&G Trumpsk) to produce a histogram of the events showing directly the exponential decay. A pulse generator provides the primary timing sequence for the measurement (Berkeley Nucleonics Corporation BNC 8010).

We take sets of data for about 1500 s, that are individually processed, and fitted. The total number of counts in a set is typically in the order of \( 3 \times 10^5 \). Figure 3 shows the accumulated decay of a set of data, together with the exponential fit and the residuals.

We apply a pile-up correction, that accounts for the preferential counting of early events. As low count rates keep this correction small, we collect data with a small number of fluorescence photons. We typically count one photon every 500 cycles. The correction alters the fitted lifetime by +0.1%. We perform a nonlinear least square fit and use an iterative algorithm to find the fitting parameters that produce the smallest \( \chi^2 \).
The decay signal $S_{8s}$ through the $7P_{3/2}$ state is a sum of two exponentials $A_B$ and a background $A_B$ with a slope $A_S$, from the turn off of the trap. The fitting function is:

$$S_{8s} = A_B + A_S t + A_{8s} \exp\left(\frac{t}{\tau_{8s}}\right) + A_{7p} \exp\left(-\frac{t}{\tau_{7p}}\right),$$

where $\tau_{7p}$ is the known lifetime of the $7P_{3/2}$ state and $\tau_{8s}$, $A_B$, $A_S$, and $A_{7p}$ are the fitting constants. Figure 3 shows an example of a data set and the fit. We start the fit 20 ns after both excitation lasers are turned off. The fitting function describes the data well, and the reduced $\chi^2$ of this particular decay is 1.11. A discrete Fourier transform of the residuals shows no structure. The average $\chi^2$ for all the data files used to obtain the lifetime is $1.07 \pm 0.07$. A change (within our quoted uncertainty) on the calibration of the linearity of the MCA is responsible for deviation from unity of the reduced chi squared. The slope that we find is 0.02 counts in 500 channels for a counting time of 1 second and it is the remanent of the trap light. A fit to a file consisting of the sum of all files gives consistent results both for the $8s$ lifetime and for the $7P_{3/2}$ lifetime when this last one is left as a free parameter.

We calculate the contribution to the uncertainty in the $8s$ lifetime from the $7P_{3/2}$ lifetime of 21.02(11) ns using Bayesian statistics. The $7P_{3/2}$ state gives a Bayesian error of 0.15%.

We do not observe any systematic effects depending on the start and end points of the fit, the so called truncation error, beyond the statistical uncertainty. We look for effects in the lifetime from imperfect lasers turn off by leaving the first photon on continuously. The change in the lifetime with the first photon off or continuously on during the decay constraints the uncertainty from imperfect lasers turn off to $\pm 0.07\%$. The time calibration of the pulse detection system contributes $\pm 0.01\%$ to the uncertainty. The TAC and MCA nonuniformity contribute $\pm 0.11\%$ error in the $8s$ level lifetime and increases the value of the $\chi^2$.

We study the effect of the initial state conditions on the obtained lifetime by changing external parameters of the measurement. We vary the power of the $817$ nm first photon laser and we observe no change in the measured lifetime. The time of flight of the atoms can influence the measured $8s$ level as excited atoms may leave the imaging region before they fluoresce. However, the average velocity of the atoms in the MOT is less than 0.1 m/s and the imaging region has a diameter of 1 mm. The time it takes the atoms to traverse the imaging region is approximately $10^3$ times the measured $8s$ level lifetime.

The slope in the fitting function influences the value of the obtained lifetime. Files with and without the second photon that produces the background exponential decay give a consistent slope. We compare the lifetime obtained by leaving the slope as a free parameter or by fixing it to the background files value and obtain an uncertainty contribution of $\pm 0.36\%$.

The counting PMT is continuously on and detects light from both the two photon excitation and the fluorescence light from the MOT. We bound the possible saturation effects on the PMT by comparing the average response of the PMT in photon counting mode with the response of a fast photodiode not subject to saturation. We find a maximum contribution of $\pm 0.24\%$ to the overall uncertainty from the PMT recovery.

We search for other possible systematic effects in the lifetime of the equivalent level ($6s$) in Rb, given the complications of working with Fr. These measurements are performed both in a vapor cell and in a MOT. There can be collisional quenching or radiation trapping in a gas of atoms that can modify the lifetime; however, we find no evidence of change when we vary the number of atoms from $10^4$ to $10^5$ in the Rb trap and we establish a limit on radiation trapping from the Rb data of $\pm 0.01\%$. We have performed an extensive search for some additional magnetic sensitivity: there is no change in the lifetime beyond the statistical uncertainty when we change the gradient of the Fr MOT. The detection of the cascaded photon reduces the possibility of quantum beats. We establish a limit on magnetic field effects of $\pm 0.11\%$ in the uncertainty of the Fr measurement consistent with our work in Rb.

Table I contains the error budget for the $8s$ level life-
time measurement. The statistical error dominates the uncertainty of the measurement. We obtain a lifetime of 53.30 ± 0.44 ns for the 8s level of francium.

Figure 4 compares the obtained 8s level lifetime with theoretical calculations. a to d are ab initio MBPT calculations of the dipole matrix elements by a: Safronova et al. [6], b: V. A. Dzuba et al. [8], c: W. R. Johnson et al. [10], and d: V. A. Dzuba et al. [11]. We calculate the lifetime with Eq. 1 from their predictions and measured transition energies. e to h are semiempirical calculations: e: M. Marinescu et al. [18], f: C. E. Theodosiou [19], g: E. Biémont et al. [20], and h: W. A. van Wijngaarden et al. [21]. The scatter of results from the MBPT calculations is small and they are all within one percent of our result. The semiempirical methods are less accurate and they have a broader scatter for their predictions (expanded scale in Fig. 4).

Our measurement establishes that the MBPT calculations of matrix elements that contribute to the total lifetime of the state are very good. They take into account the large relativistic effects present in this heavy atom as well as the multiple correlations from its 87 electrons. Their accuracy is vital for future interpretation of PNC measurements. The agreement of theoretical predictions over different species reinforces the interpretation of PNC measurements in Cs which are now sensitive to the nuclear weak force. [11].

Work supported by NSF. E. G. acknowledges support from CONACYT and the authors thank the personnel of the Nuclear Structure Laboratory at Stony Brook for their support as well as J. Gripp, J. E. Simsarian and B. Minford for equipment loans.

TABLE I: Error budget for the 8s level lifetime measurement.

| Error [%] |
|-----------------|
| Time calibration | ±0.01 |
| Bayesian error | ±0.15 |
| TAC/MCA response nonuniformity | ±0.11 |
| Radiation trapping | ±0.01 |
| Imperfect laser turnoff | ±0.07 |
| Magnetic Field | ±0.11 |
| Background slope | ±0.36 |
| PMT response | ±0.24 |
| Statistical error | ±0.65 |
| **Total** | ±0.82 |

FIG. 4: Comparison of the 8s atomic lifetime (ns) with theoretical calculations. The statistical error dominates the measurement. The calculations are labelled with letters explained in the text and with numbers that indicate the reference.

![Diagram showing comparison of 8s atomic lifetime with theoretical calculations](image)