Lifetime of the $^2F_{7/2}$ level in Yb$^+$ for spontaneous emission of electric octupole radiation

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(Dated: July 26, 2021)

We report a measurement of the radiative lifetime of the $^2F_{7/2}$ level of $^{171}$Yb$^+$ that is coupled to the $^2S_{1/2}$ ground state via an electric octupole transition. The radiative lifetime is determined to be $4.98(25) \times 10^7$ s, corresponding to 1.58(8) years. The result reduces the relative uncertainty in this exceptionally long excited state lifetime by one order of magnitude with respect to previous experimental estimates. Our method is based on the coherent excitation of the corresponding transition and avoids limitations through competing decay processes. The explicit dependence on the laser intensity is eliminated by simultaneously measuring the resonant Rabi frequency and the induced quadratic Stark shift. Combining the result with information on the dynamic differential polarizability permits a calculation of the transition matrix element to infer the radiative lifetime.

Coherent interrogation of trapped particles facilitates the determination of atomic transition frequencies with high accuracy, recently demonstrated below the $10^{-18}$ fractional uncertainty level [1]. Complementary information on the electronic structure of atomic systems can be obtained from measurements of coupling strengths between electronic states and of their natural lifetimes. For transitions with coupling via optical electric dipole (E1) radiation, excited states show natural lifetimes of nanoseconds that can be determined directly with a relative uncertainty at the $10^{-3}$ level by observing the spontaneous decay (see for example [2, 3]). Even higher precision has been obtained by using theoretical information on the atomic structure together with measurements of dynamic Stark shifts resulting from the E1 coupling to Rydberg states [4]. As recently demonstrated, by combining measurements of transition rates and Stark shifts from the same laser, an explicit dependence on its intensity is avoided and the E1 coupling strengths can be determined precisely without theoretical modeling [5]. Low-lying electronic states for which decay is not supported by E1 selection rules show significantly longer natural lifetimes. Such meta-stable states of isolated quantum systems provide the basis for application in frequency metrology, quantum simulation, and quantum information processing [6, 7].

If the natural lifetime exceeds seconds, a direct observation of the spontaneous decay is particularly challenging because of competing processes such as collisions with the background gas, off-resonant laser radiation, and the long measurement periods needed to achieve sufficiently small statistical uncertainty [8,9]. Using large ensembles of laser-cooled neutral atoms confined in magnetic quadrupole traps, it has been possible to investigate excited state lifetimes exceeding minutes [10,11]. With this technique, a lifetime $\tau = 7870(510)$ s has been found for the $^2S_{1/2}$ state of Helium, to our knowledge the longest natural lifetime on an optical transition determined experimentally so far [12]. For even longer lifetimes, the competing processes dominate and make a direct measurement of the natural decay time increasingly difficult.

In this Letter, we devise an alternative method to determine the natural lifetime of metastable states for systems with a single radiative decay channel from an excited state $|e\rangle$ to a ground state $|g\rangle$. The method relies on monitoring the coherent time evolution of the two-level system while it is resonantly excited by a laser, in order to extract the matrix element of the transition. To illustrate the method, we investigate the $^2S_{1/2}(F = 0) \rightarrow ^2F_{7/2}(F = 3)$ electric octupole (E3) transition in a single trapped $^{171}$Yb$^+$ ion and infer the yearslong natural lifetime of the excited $^2F_{7/2}$ state with 5% uncertainty.
As shown in Fig. 1, our result reduces the relative uncertainty by one order of magnitude with respect to previous long-standing estimates [13, 15] and provides a clear point of reference for atomic structure calculations.

The method presented here is based on the fact that the radiative lifetime \( \tau \) is related to the matrix element \( V_{eg} \) of the transition according to Fermi’s golden rule, \( \tau \propto 1/|V_{eg}|^2 \), where the proportionality coefficient depends on the experimental geometry and the angular momentum of ground and excited state. If the transition is coherently driven by a laser, the matrix element is measurable from the Rabi frequency \( \Omega \) describing the oscillation of population between \( |g\rangle \) and \( |e\rangle \) states: \( \Omega = 2\pi E_0|V_{eg}|/\hbar \), with \( \hbar \) the Planck constant. Independence from the electric field amplitude \( E_0 \) of the laser at the ion position can be achieved by simultaneously measuring the differential quadratic Stark shift \( \Delta \nu_{QS} = E_0^2\alpha_{eg}(\nu_0)/(2\hbar) \) and defining the relative excitation strength \( \xi = \Omega^2/\Delta \nu_{QS} \). This quantity together with the differential polarizability \( \Delta \alpha_{eg} \) at the transition frequency \( \nu_0 \), obtained independently from experiments or theory, permits the determination of the matrix element: \( |V_{eg}| = 1/(2\pi)^\nu \hbar \xi \Delta \alpha_{eg}(\nu_0)\Omega^2/2 \).

The Yb\(^+\) E3 transition is employed in optical atomic clocks [20, 21] and currently features the most accurately determined transition frequency [22]. It is well-suited for various searches for physics beyond the standard model [23] and has been used to realize the most stringent limits for potential violations of local Lorentz invariance in the electron sector [21]. Because of the large sensitivity of the transition frequency to variations of the fine structure constant \( \alpha \), repeated comparisons to other frequency references currently provide the most rigorous constraints on a temporal variation of \( \alpha \) and a coupling of \( \alpha \) to gravity [22]. Besides applications in other searches for new physics [24, 25], Yb\(^+\) is employed in a number of quantum computing experiments [26, 28], which can take advantage of the long-living \( ^2F_{7/2} \) state [27, 29].

In our experiment, a single ion is confined in a radio frequency Paul trap at ultrahigh vacuum and laser-cooled on the \( ^2S_{1/2} \rightarrow ^2P_{1/2} \) transition at 370 nm close to the Doppler limit. During cooling, population trapping in the \( ^3D_{5/2} \) state is prevented using laser radiation at 935 nm. Stray electric fields are compensated to suppress the relative strength of first-order micromotion sidebands to less than 1%. The frequency of the 467 nm (642 THz) probe laser is stabilized via a frequency comb generator to the length of a cryogenic silicon cavity [30] and permits coherent excitation of the E3 transition with laser pulses of up to 500 ms duration. After successful excitation, the \( ^2F_{7/2} \) state can be rapidly depopulated using laser radiation at 760 nm. Without this repump laser, the \( ^2F_{7/2} \) state is quenched by collisions with the residual gas [31, 32] and via excitation of the \( ^2F_{7/2} \rightarrow ^2D_{5/2} \) transition by room-temperature thermal radiation, and a lifetime of several hours is typically observed. The linear polarization \( \epsilon \) of the probe laser beam with wave vector \( \mathbf{k} \) and the orientation of the ion quantization axis, defined by an externally applied magnetic field \( \mathbf{B} \), are chosen to maximize the excitation probability: We align \( \epsilon \), \( \mathbf{k} \) and \( \mathbf{B} \) within one plane and set the angle \( \beta_{E3} \) between \( \epsilon \) and \( \mathbf{B} \) to 59(1)° [33]. To derive the resonant Rabi frequency, excitations of the E3 transition with variable pulse duration are performed. Under ideal conditions with the ion in the motional ground state, the Rabi frequency can be directly deduced from the oscillation of the excitation probability, \( p(t) = (1 - \cos(\Omega t))/2 \). However, in our experimental realization with a Lamb-Dicke parameter of about 0.08, Doppler cooling to a mean motional quantum number of about 30 leads to considerable damping of the oscillation due to the different couplings between ground and excited motional states [34] as shown in the inset of Fig. 2. Taking this into account, the Rabi frequency is determined for various settings of the probe laser intensity. For each intensity the quadratic Stark shift is measured as the offset \( \Delta \nu_{QS} \) from the unperturbed transition frequency \( \nu_0 \). In order to determine \( \nu_0 \) and to correct for the frequency drift of the silicon reference cavity, the measurements are complemented by periods where the experiment is operated as an optical clock so that the laser frequency is locked to \( \nu_0 \) [22]. Thereby, \( \Delta \nu_{QS} \) is measured with a relative uncertainty of less than 1%. The results are depicted in Fig. 2. Assuming a linear dependence for the data, we obtain the relative excitation strength \( \xi = 30.3(9) \) Hz. The uncertainty results from statistics, the residual motion of the ion and possible deviations from the resonant frequency during the determination of the Rabi frequency.

In addition to the relative excitation strength for the E3 transition, the differential polarizability needs to be
The obtained value for the lifetime is compared to previous measurements and calculations in Fig. 1. Our result reduces the relative uncertainty by about one order of magnitude with respect to previous long-standing estimates. It represents to our knowledge the longest experimentally determined natural lifetime of an electronic state to date and the first precise measurement of an E3 radiative lifetime. The achieved relative uncertainty of 5% is mainly limited by the residual temperature of the trapped ion in the measurement of the relative excitation.
strength and by the determination of the differential polarizability. While the former contribution can be easily reduced by resolved sideband cooling \cite{9}, superior accuracy in the differential polarizability is obtained for other ion species such as \(^{88}\text{Sr}^+\) \cite{10}, \(^{40}\text{Ca}^+\) \cite{11}, or \(^{138}\text{Ba}^+\) \cite{12}. Co-trapping \(^{171}\text{Yb}^+\) with such an ancillary ion permits a transfer of the relative accuracy in the differential polarizability.

Our experimental result for the lifetime has been obtained with \(^{171}\text{Yb}^+\) with nuclear spin \(I = 1/2\) while earlier work has also investigated \(^{172}\text{Yb}^+\) with \(I = 0\) \cite{13} (see Fig. 1). For the \(^{173}\text{Yb}^+\) isotope with nuclear spin \(I = 5/2\) a shortening of the lifetime of some of the hyperfine levels of the \(2\text{F}_3/2\) state by more than 2 orders of magnitude has been predicted \cite{14}. The effect is due to the admixture of the \(2\text{P}_{3/2}\) state that is induced by hyperfine interaction with the nuclear electric quadrupole moment. The lifetime of the unperturbed E3 decay that we have determined here is expected to be valid for all stable isotopes of Yb with the exception of \(^{173}\text{Yb}^+\) and provides the reference for a quantitative experimental study of the hyperfine quenching effect in the latter \cite{15}.

The method of combining measurements of the resonant Rabi frequency and the quadratic Stark shift for the determination of a small transition matrix element is readily applicable to other atomic species featuring electronic states with natural lifetimes exceeding minutes and promises high accuracy due to the immunity to competing decay processes. Particularly for highly-forbidden transitions in \(^{175}\text{Lu}^+\) \cite{16}, \(^{40}\text{Ca}^+\) \cite{17}, alkaline-earth atoms \cite{18}, and highly-charged ions \cite{19} that are of interest for optical clocks and searches for violations of fundamental symmetries, accurate assessments of the lifetime support a better understanding of the atomic structure.

We thank Burghard Lipphardt, Thomas Legero, Erik Benkler and Uwe Sterr for providing the ultrastable laser reference Si-2. This work has been supported by the Max-Planck-RIKEN-PTB-Center for Time, Constants and Fundamental Symmetries. Furthermore, this work has been funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) - Project-ID 274200144 - SFB 1227 within project B02 and under Germany’s Excellence Strategy - EXC-2123 Quantum Frontiers - 390837967.
