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Relativistic Electron Correlation, Quantum Electrodynamics, and the Lifetime of the $1s^2 2s^2 2p\,^2P^o_{3/2}$ Level in Boronlike Argon

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The lifetime of the Ar^{13+} $1s^2 2s^2 2p\,^2P^o_{3/2}$ metastable level was determined at the Heidelberg Electron Beam Ion Trap to be 9.573(4)(5) ms(stat)(syst). The accuracy level of one per thousand makes this measurement sensitive to quantum electrodynamic effects like the electron anomalous magnetic moment (EAMM) and to relativistic electron-electron correlation effects like the frequency-dependent Breit interaction. Theoretical predictions, adjusted for the EAMM, cluster about a lifetime that is approximately 3σ shorter than our experimental result.

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Increasingly accurate measurements of transition energies and excited-state lifetimes in few-electron systems represent a challenge to theory because of the complex interplay of relativistic, correlation, and quantum electrodynamic (QED) effects. At medium nuclear charge Z, the effects are all intertwined. For instance, the nuclear charge number of argon ($Z = 18$) is close to a "phase transition" occurring near $Z \approx 26$ where the spin-orbit coupling [a relativistic effect of the order of $(Z\alpha)^4 mc^2$] exceeds the interelectron Coulomb interaction (order $Z\alpha^2 mc^2$). Here, α is the fine-structure constant, and mc^2 is the electron rest mass energy. Measurements of excited-state lifetimes are particularly needed since they are especially sensitive to the long-range behavior of the electronic wave function.

Here, we present an accurate lifetime measurement of the $1s^2 2s^2 2p\,^2P^o_{3/2}$ metastable level of boronlike Ar^{13+}. Three previous measurements [1–3], although quite recent, were more than an order of magnitude less accurate than our measurement of 9.573(4)(5) ms(stat)(syst), which is in agreement with the latest result of 9.70(15) ms [3].

The measurement reported here [4] is sensitive to the relativistic correlation as well as the electron anomalous magnetic moment (EAMM) (dominant QED effect), which shortens the lifetime of this metastable level by a relative factor $1 - 2\alpha/\pi$, as a straightforward calculation based on the well-known effective Dirac equation (see, e.g., Chap. 7 of [5]). Seemingly, our measurement is in excellent agreement with most theoretical calculations reported in the literature [6–14], which do not include the EAMM effect. Surprisingly, the agreement disappears if one adjusts the theoretical calculations by the EAMM contribution (which should by all means be included in theory at the current level of experimental accuracy), and most theoretical predictions appear to cluster about a value for the lifetime (9.53 ms) that is approximately 3σ shorter than our experimental result. While we cannot solve the puzzle, we would like to emphasize a new theoretical approach that might resolve the disagreement.

The measurement was performed at the Electron Beam Ion Trap (EBIT) of the Max-Planck-Institut für Kernphysik [15] (schematically drawn in Fig. 1). The EBIT was operated at effective electron beam energies of 700 and 715 eV at the trap center (first and second series of measurements [16], respectively). This energy was just sufficient to ionize Ar^{12+}, but not Ar^{13+}, as their respective ionization potentials are 686 and 755 eV [17]. The beam current was typically set to 100 mA. The two drift tubes closest to the trap center were used to confine ions axially by applying an electrostatic potential of 1.5 kV, which suppressed axial ion losses. The trap was dumped every 10 s, for 100 ms, to avoid the accumulation of heavy contaminant ions.

The lifetime of the $1s^2 2s^2 2p\,^2P^o_{3/2}$ level was measured by monitoring its decay to $1s^2 2s^2 2p\,^2P^o_{1/2}$ through an M1 transition [$\lambda = 441.2559(1)$ nm; see Ref. [18]] in the

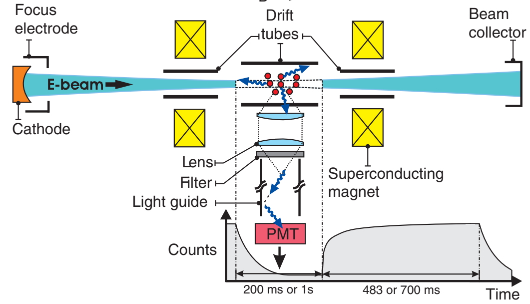

FIG. 1 (color online). Experimental setup at the MPI-K EBIT.

magnetic trapping mode [19]. This was done by cyclically turning the electron beam off for 200 ms or 1 s and on for 483 or 700 ms, by applying negative or positive high voltage to the focus electrode of the electron gun. Photons emitted by the trapped ions were collected by an $f/4$ lens ($\oslash = 38$ mm, $f = 150$ mm) located inside the EBIT, and led to a cooled photomultiplier tube (PMT) with a dark count rate of about 30 counts/s, by means of a special 1150-mm long light guide. The large field of view of the lens, the wide acceptance angle of the light guide, and the width (7 mm) of the center drift tube slits allowed to collect light from the trapped ions without any loss of efficiency even as the ion cloud slowly expands while the beam is off, avoiding possible systematic errors caused by metastable ions not seen by the light collection system. The Ar^{13+} spectral line was selected by an interference filter with a transmission of 60% at 442 nm and a 3 nm bandwidth (FWHM), efficiently rejecting the cathode glow.

In total, decay curves for various experimental conditions were obtained for 116 hours. Typical examples are shown in Fig. 2. In addition to the 10 ms decay (t_1) of the metastable level, a background contribution (decay time $t_2 \approx 1$ s) is observed. Beginning ca. 2 ms after turning off the electron beam, the signal $N(t)$ can be fitted to a linear combination of two exponentials: $N(t) = a_1 \exp(-t/t_1) + a_2 \exp(-t/t_2) + y_o$, where a_1, t_1, a_2, and t_2 are free parameters, and y_o is the PMT dark count rate. Other fit functions using three or more exponentials failed to yield a satisfactory representation of the data. The χ^2/(degree of freedom) as well as the R^2 obtained by the preferred fitting procedures were on average 1.15 and 0.999, respectively. A standard tail-fit analysis performed by truncating the start and end times in the fitting showed changes consistent with statistical fluctuations. As an example, for a high statistics (binned) decay curve taken for a total time of 960 min (first series) we measured 9.573(4), 9.571(6), 9.573(8), and 9.570(10) ms, by truncating the start time to 2.1, 7.1, 12.1, and 17.2 ms, respectively.

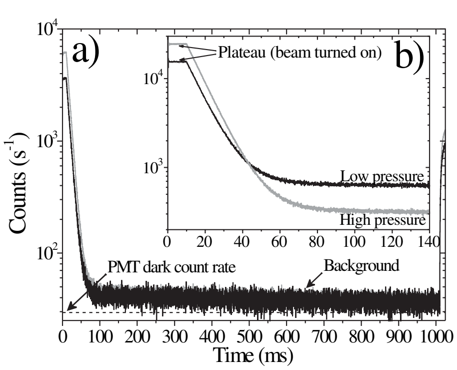

FIG. 2. Typical time-normalized decay curves for two series of measurements taken at different injection gas pressures, as reflected in the heights of the plateaus: (a) is for 1 s beam off time (111.1 μs/channel), and (b) is for 200 ms beam off time (50 μs/channel).

The dead time $t_d = 1.4(1)$ μs of the data acquisition system was taken into account by the relation $N_c = N(t)/[1 + N(t)t_d/t_a]$, where N_c is the number of photons counted by the data acquisition system, and t_a is the total acquisition time per channel.

The time-averaged height of the slowly decaying component, hereafter denoted as the background level, similarly observed in a previous experiment [20], could, in principle, be caused by four physical phenomena: (i) ion-ion collisional excitation, (ii) charge-exchange collisions of neutral atoms with Ar^{14+} ions, (iii) cascade repopulation, and (iv) electron-impact excitation of Ar^{13+} ions. By extensive systematic measurements at different gas pressures, for various electron beam currents, for very long (1 s) observation time windows and different trapping potentials along with quantitative arguments, we can rule out all but the last of these mechanisms. While full details will be presented in a forthcoming paper, we have in short the following facts: the overall decrease of the background level for high injection gas pressures [see Fig. 2(b)] immediately discards (i) and (ii). Process (ii) is also excluded since the electron beam energy at the center of the trap is still too low to produce Ar^{14+} ions. Reassuringly, even as the beam energy was intentionally raised to 850 eV, no effect could be observed. Cascade repopulation (iii) is discarded based on (a) quenching of high-lying levels by field ionization and Stark l mixing [21] induced by the electron beam, and (b) the relatively short average lifetime of highly excited Rydberg states for the medium-Z ion under investigation [22], which is less than a few μs and not a one-second time scale. This latter argument is also supported by atomic structure calculations dedicated to B-like ions [23].

Impact excitation by low-energy electrons (iv) trapped by the ion-cloud space charge potential (SCP) (see also Ref. [24]) can be estimated as follows. The difference between the total experimental SCP and the theoretical electron beam SCP (-550 V at the electron beam radius) yields an ion SCP of 45% of the electron beam SCP, i.e., $+250$ V. The temperature T_e of the trapped electrons can be estimated from the relation $\omega = qV/(k_B T_e)$, where q is the elementary charge, V the trapping potential (ion SCP), and k_B denotes the Boltzmann constant. Typically, ω takes values in the range 2 to 10 (see Ref. [25]) and hence $k_B T_e \approx 75$ eV. Additionally, the ion temperature T_i was determined to be $k_B T_i = 350$ eV from the measured Doppler width of the spectral line. The density of trapped electrons can be extracted from the background count rate using calculated Maxwellian-averaged rate coefficients [26]. It is found to be around 5×10^7 cm^{-3}, approximately 60 times less than the ionic density of 3×10^9 cm^{-3}, which was estimated from the yield of extracted ions in an independent experiment. With the above-mentioned parameters, simulations using the Spitzer equation [27] show that, although the trapped electrons cool down through synchrotron radiation with a time constant of 56 ms in the 8 T magnetic field, they keep a relatively constant temperature of $T_e \approx 50$ eV, sufficient to excite the transition, over more than 1 s, consistent with the mechanism (iv).

Because of the presence of trapped electrons, the determination of the lifetime has to include the effect of collisional deexcitation to the ground state and excitation to higher levels. Neglecting any losses out of the trap, the time evolution of the population of the metastable level can be described as [28]

$$N_M(t) = \left[N_M(0) - \frac{N_t k n_e}{1/\tau + 2kn_e}\right]\exp\left[-\left(\frac{1}{\tau} + 2kn_e\right)t\right] + \frac{N_t k n_e}{1/\tau + 2kn_e}, \qquad (1)$$

where $N_M(0)$ is the initial number of metastable ions before turning off the electron beam, τ is the lifetime, N_t is the total number of Ar^{13+} ions, k is the excitation or deexcitation rate coefficient, and n_e is the electron density. The quantity $t_1 = [1/\tau + 2kn_e]^{-1}$ is an apparent lifetime, and the last term in Eq. (1) describes a constant background level. Multiplying the latter with the detection efficiency and the atomic transition probability $1/\tau$, and assuming $N > 5 \times 10^7$ stored ions in the trap (estimated from ion extraction), an upper limit of the quenching rate can be obtained by comparison to the maximum experimental background rate (see Fig. 2), and it is found to be less than 0.3% of the atomic transition probability. This figure is, however, a gross overestimate: quenching depends on the number of trapped electrons and ions, and if it were important, then it should manifest itself in a systematic dependence of t_1 on the ratio of the background-to-plateau level, which is a normalized measure of the number of trapped electrons. As demonstrated in Fig. 3, where t_1 is plotted as a function of this ratio, one cannot find such a correlation. This holds also for correlations of t_1 versus the height of the plateau (absolute count rate) and the height of background and t_2. Reassuringly, we found that when the electron beam was only partially turned off (to a residual current a few mA), we were able to suppress the background level down to the PMT dark count rate, the explanation being that collisions with beam electron efficiently heat up and expel low-energy electrons out of the trap region. The background of the first series of measurements was typically ca. 3 times higher than that of the second series, but the measured lifetimes are essentially the same (see open and solid circles in Fig. 3).

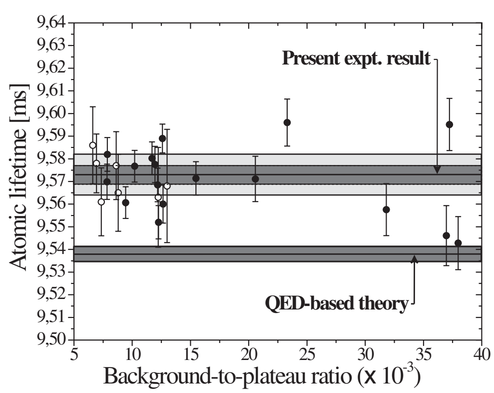

FIG. 3. Lifetimes from decay curves (white circles: second series) taken for different run times (4–5 hours per data point) as a function of the ratio of the background level to the height of the plateau observed before turning off the electron beam [see Fig. 2(b)]. Our experimental value is compared to a CIDFS prediction [9].

Directly after turning off the electron beam, short-time ion losses by (a) the dump, (b) electron-ion recombination, (c) charge-exchange recombination, and (d) evaporative cooling may reduce the Ar^{13+} population. None of these processes influence our lifetime measurements on the level of the final uncertainty, except for the dump-induced loss rate of 10^{-2} s^{-1}, for which our measurements are corrected. Losses by electron-ion recombination are discarded because for $T_e \approx 50$ eV and an electron density of 5×10^7 cm^{-3}, the Maxwellian-averaged recombination rate is estimated to be of the order of 10^{-4} s^{-1}. Charge-exchange losses are negligible because experimental investigations at injection pressures varying by 1 order of magnitude yielded no discernible reduction of the lifetime, in accordance with our estimates. This experimental observation is in agreement with a Z scaling of charge-exchange rate measurements carried out at the Lawrence Livermore EBIT with bare Kr^{36+}, which survives in the magnetic trapping mode [19] for at least 5 s. Evaporative cooling was investigated by measuring t_1 and t_2 as a function of the trapping potential, which was varied from 0.1 up to 2.5 kV. Whereas a distinct increase of t_2 and also the background height were observed, no change in t_1 for trapping potentials of more than 500 V was found. To support this, the axial ion escape rate out of the trap can be estimated based on an approximation of the Fokker-Planck equation [25]. For an ion temperature of $k_B T_i = 350$ eV, charge $q = 13$, and the drift tube at $V = 1.5$ kV, we have ω close to 50, and therefore the axial ion loss rate can be neglected. Changes in the background decay times (t_2) are then due to losses of electrons trapped by the ion-cloud SCP. According to the discussion above, the only remaining systematic error (that we are aware of) to our experimental lifetime is the error bar of the measured total data acquisition dead time. This introduces a 0.005 ms uncertainty that was added linearly to the statistical error.

Our experimental result is $\tau = 9.573(4)(5)$ ms(stat) (syst). A comparison with previous theoretical and experimental results is shown in Fig. 4 (theory: MCDF [6], MCBP [7], C.-S [8], CIDFS [9], SS'98 [10], RQDO [11]

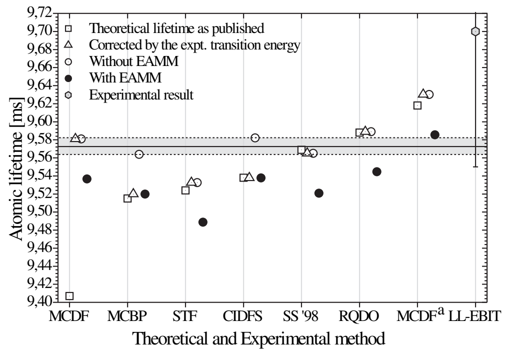

FIG. 4. Theoretical lifetimes [6–14] and (rightmost) the experimental result of Ref. [3]. The EAMM contribution had been ignored in all theoretical calculations except for Refs. [7,9].

and MCDF^a [12]; experiment: Ref. [3]). A recent calculation using the configuration-interaction Dirac-Fock-Sturm method (CIDFS, Ref. [9]) leads to a theoretical value of $\tau = 9.538$ ms. The decay rate depends on the cube of the transition wavelength that has been measured accurately in a recent experiment [Ref. [18], $\lambda = 441.2559(1)$ nm]. The theoretical calculations (except [9]) did not have this information at their disposal, and, thus, in order to give a more complete representation of the theoretical data, theory is plotted in various ways: (1) as published, (2) corrected for the experimental transition wavelength, and (3) excluding and (4) including the contribution due to the EAMM, which leads to a decrease of the lifetime by a factor of $1 - 2\alpha/\pi$. It is difficult to interpret the obvious scatter of the theoretical calculations, which appear to cluster around a lifetime of 9.53 ms, in a 3σ disagreement with our experiment.

In typical relativistic many-body calculations like the multiconfiguration Dirac-Fock (MCDF) method, one usually starts from fully relativistic one-particle orbitals. The electron-electron correlation, as well as relativistic correlation effects (like the frequency-dependent part of the Breit interaction, which leads to a 1.5 per mille shift of the decay rate) and QED are added on later. Very recently, a completely different approach has been proposed where the sequence of approximations is reversed: it is based on effective many-body Hamiltonians and nonrelativistic quantum electrodynamics (NRQED; see Ref. [29]). In this approach, the correlation effects are taken into account to an essentially arbitrary accuracy right from the start of the calculation. Typically, one can calculate relativistic and radiative corrections to decay rates up to the relative order of $\alpha(Z\alpha)^2$ by NRQED methods [30], easily sufficient for a relative accuracy of 10^{-4} at $Z = 18$.

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