

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

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(Dated: June 3, 2005)

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

PACS numbers: 32.70.Cs, 31.30.Jv

Many-electron systems at medium nuclear charge  $Z$  represent a challenge to theory because of the interplay of relativistic, correlation, and quantum electrodynamic (QED) effects, which are all intertwined. In particular, the nuclear charge number of argon ( $Z = 18$ ) is close to a “transition range” where relativistic effects [spin-orbit coupling, order  $(Z\alpha)^4 mc^2$ ] become larger than the  $1/r_{12}$ -Coulomb interaction (which is of order  $Z\alpha^2 mc^2$ ). Here,  $\alpha$  is the fine-structure constant, and  $mc^2$  is the electron rest mass energy. The transition occurs near  $Z \approx 26$ . Measurements of decay rates are especially sensitive to the detailed structure of the electronic wave function which enters the transition matrix element, typically considerably harder to calculate than the energy eigenvalue, and thus represent stringent benchmarks.

Here, we present an accurate lifetime measurement of the  $1s^2 2s^2 2p^2 P_{3/2}^{\circ}$  metastable level of  $\text{Ar}^{13+}$  (M1 decay). Two previous measurements [1, 2], although quite recent, were more than an order of magnitude less accurate than our result of  $9.573(4)(5)$  ms (stat)(syst). Our value is in agreement with Ref. [3], where the lifetime had been obtained as  $9.70(15)$  ms. However, the accuracy of the latter experiment was still insufficient for a verification of high-precision atomic theory (relativistic correlation and QED effects) [5].

The measurement reported here is sensitive to the relativistic correlation as well as the dominant QED contribution (EAMM) which shortens the lifetime by a relative factor  $1 - 2\alpha/\pi$ , as a straightforward calculation based on the well-known effective Dirac equation shows (see, e.g., Chap. 7 of [4]). Seemingly, our measurement is in excellent agreement with various theoretical calculations reported in the literature [6–14], which do not include the EAMM contribution. Surprisingly, the agreement disappears if one adjusts the theoretical calculation by the EAMM contribution (which should by all means be included in theory at the current level of experimental accuracy), and various theoretical calculations appear to cluster about a value for the lifetime that is about  $3\sigma$  shorter than our experiment. The EAMM had been ignored in

most theoretical approaches, most likely because the authors had not aimed for the level of accuracy that can now be obtained experimentally. While we cannot resolve the resulting puzzle, we would like to emphasize that steps toward a clarification might involve novel theoretical concepts introduced very recently in the context of higher-order effective Hamiltonians for (arbitrary) light atomic systems.

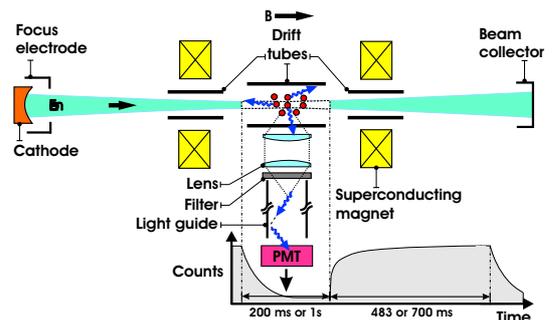


FIG. 1: Experimental set-up at the MPI-K EBIT.

The measurement reported in this Letter 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 eV and 715 eV at the trap center (first and second series of measurements, respectively). This energy was just sufficient to ionize  $\text{Ar}^{12+}$ , but not  $\text{Ar}^{13+}$ , as their respective ionization potentials are 686 eV and 755 eV [16]. An additional measurement at 850 eV tested the effect of  $\text{Ar}^{14+}$  depletion. 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 escape. The trap was dumped every 10 s to avoid the accumulation of heavy contaminant ions.

The lifetime of the  $1s^2 2s^2 2p^2 P_{3/2}^{\circ}$  level was measured by monitoring its decay to  $1s^2 2s^2 2p^2 P_{1/2}^{\circ}$  through an M1 tran-

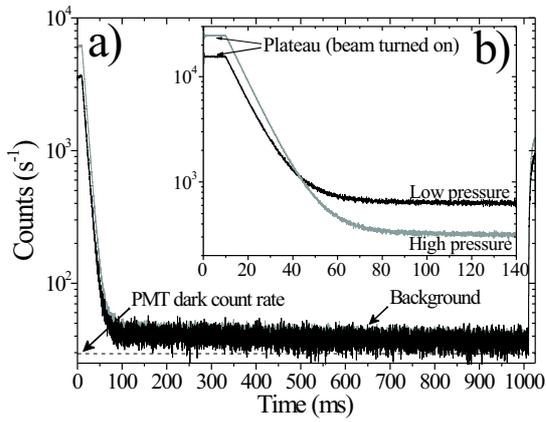


FIG. 2: Typical time-normalized decay curves for two series of measurements taken at different injection gas pressures, as reflected in the heights of the plateaus: Fig. **a**) is for 1 s beam off time (111.1  $\mu\text{s}/\text{channel}$ ), and Fig. **b**) is for 200 ms beam off time (50  $\mu\text{s}/\text{channel}$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

sition [ $\lambda = 441.2559(1)$  nm, see Ref. [17]] in the magnetic trapping mode [18]. This was done by cyclically turning the electron beam off for 200 ms or 1 s and on for 483 ms 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 ( $\varnothing = 38$  mm,  $f = 150$  mm) located inside the EBIT, 150 mm away from the central drift tube, and led to a cooled photomultiplier tube (PMT) with a dark count rate of about 30 counts per second by means of a special 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  $\text{Ar}^{13+}$  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  $\chi^2/\text{DOF}$  (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 fluctuations consistent with the statistical uncertainties of the individual data points. As an example, for a high statistics (binned)

decay curve taken for a total time of 960 minutes (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 15.2 ms, respectively.

The dead time  $t_d = 1.4(1)$   $\mu\text{s}$  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 [19], could in principle be caused by four physical phenomena: (i) ion-ion collisional excitation, (ii) charge-exchange collisions of neutral atoms with  $\text{Ar}^{14+}$  ions, (iii) cascade repopulation and (iv) electron-impact excitation of  $\text{Ar}^{13+}$  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. 2b) immediately discards (i) and (ii). In addition, (i) is ruled out because for excitation to occur between such highly charged ions, their mutual Coulomb repulsion must be overcome; this would require ionic kinetic energies beyond those of the trapped ions. Process (ii) is also excluded since the electron beam energy at the center of the trap is still too low to produce  $\text{Ar}^{14+}$  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 [20] induced by the electron beam; and (b) the relatively short average lifetime of highly excited Rydberg states for the medium- $Z$  ion under investigation [21], which is of order 10  $\mu\text{s}$  and not a one-second time scale. This latter argument is also supported by atomic structure calculations dedicated to this particular ion [22].

Impact excitation by low-energy electrons (iv) trapped by the ion-cloud space charge potential (SCP) 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 inferred from the ion temperature  $T_i$ , since they are in thermal contact by Coulomb collisions.  $T_i$  is obtained via the relation  $\omega = qV/(k_B T_i)$ , where  $q$  is the ionic charge,  $V$  is the trapping potential, and  $k_B$  denotes the Boltzmann constant. Typically, the average energy of the trapped ions remains constant below the barrier potential with  $\omega$  taking values in the range 2 to 10 (see Ref. [23]). Additionally,  $T_i$  was also 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 [24] at

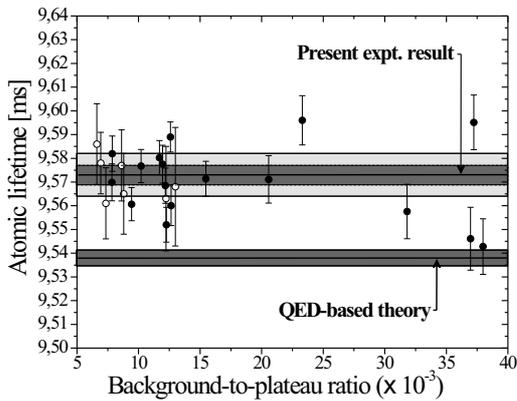


FIG. 3: Lifetimes from decay curves (white circles: second series) taken for different run times (4-5 hours per data point) as a function of the ratio of the background level to the height of the plateau observed before turning off the electron beam (see Fig. 2b). Our experimental value is compared to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

$k_B T_e \approx 75$  eV. It is found to be around  $5 \times 10^7 \text{ cm}^{-3}$ , approximately 60 times less than the ionic density of  $3 \times 10^9 \text{ cm}^{-3}$ , which was estimated from the yield of extracted ions in an independent experiment. With the above mentioned parameters, simulations applying the Spitzer equation [25] 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  $T_e \approx 50$  eV, sufficient to excite the transition, over more than 1 s, and this is consistent with the mechanism (iv).

Because of the presence of trapped electrons, the determination of the lifetime has to include the effect of collisional de-excitation 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 [26]

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

where  $N_M(0)$  is the initial number of metastable ions before turning off the electron beam,  $\tau$  is the lifetime,  $N_t$  is the total number of  $\text{Ar}^{13+}$  ions,  $k$  is the excitation/de-excitation rate coefficient, and  $n_e$  is the electron density. The quantity  $t_1 = (1/\tau + 2k n_e)^{-1}$  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  $N < 1 \times 10^7$  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 0.06% of the atomic transition probability. This figure is still an 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 white and black circles in Fig. 3) [27].

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  $\text{Ar}^{13+}$  population. None of these processes influence our lifetime measurements on the level of the final uncertainty, except for the dump-induced loss rate of  $10^{-2} \text{ s}^{-1}$ , 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  $6 \times 10^7 \text{ cm}^{-3}$ , the Maxwellian-averaged recombination rate is estimated to be of the order of  $5 \times 10^{-4} \text{ s}^{-1}$ . Charge-exchange losses are negligible because experimental investigations at injection pressures varying by one order of magnitude yielded no discernable reduction of the lifetime, in accordance with our estimates. This experimental observation is in agreement with a  $Z$ -scaling of the charge exchange rate measurements carried out at the Lawrence Livermore EBIT with bare  $\text{Kr}^{36+}$ , which survives in the magnetic trapping mode [18] 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 [23]. For an ion temperature of  $k_B T_i = 350$  eV, charge number  $q = 13$  and the drift tube at  $V = 1.5$  kV, we have  $\omega$  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 which 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] and MCDF<sup>a</sup> [12], experiment: Ref. [3]). A recent calculation using the configuration-interaction Dirac-Fock-Sturm method (CIDFS,

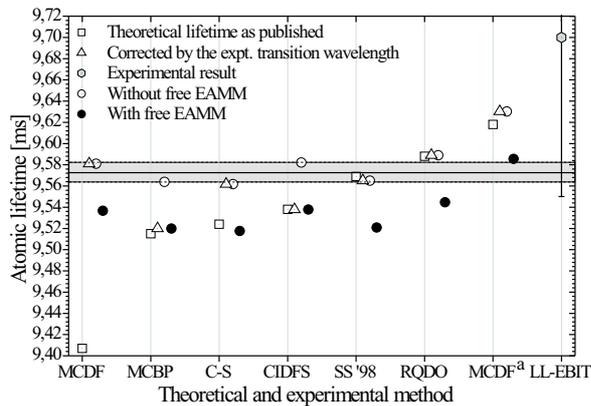


FIG. 4: Theoretical lifetimes [6–14] and (rightmost) the experimental result of Ref. [3]. The EAMM contribution had been ignored in all theoretical calculations except for Refs. [7, 9].

Ref. [9]) leads to a theoretical value of  $\tau = 9.538$  ms. The decay rate depends on the cube of the transition frequency which has been measured accurately in a recent experiment [Ref. [17],  $\lambda = 441.2559(1)$  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 [see also Eq. (3.22) of Ref. [10]], 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 about a lifetime of 9.53 ms, in a  $3\sigma$  disagreement with our experiment [28].

In typical relativistic many-body calculations like the multi-configuration 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, whose treatment has been notoriously difficult for boronlike systems, 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  $Z = 18$ .

We acknowledge support from the Max-Planck-Institut für Kernphysik. I.I.T. and A.V. acknowledge support from RFBR (Grants Nr. 01-02-17248 and 00-03-33041) and from the program “Russian Universities” (Grant Nr. UR.01.01.072).

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