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Correlation and quantum electrodynamic effects on the radiative lifetime and relativistic nuclear recoil in Ar^{13+} and Ar^{14+} ions

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Abstract.

The radiative lifetime and mass isotope shift of the $1s^2 2s^2 2p^2 P_{3/2} - ^2P_{1/2}$ $M1$ transition in Ar^{13+} ions have been determined with high accuracies using the Heidelberg electron beam ion trap. This fundamentally relativistic transition provides unique possibilities for performing precise studies of correlation and quantum electrodynamic effects in many-electron systems. The lifetime corresponding to the transition has been measured with an accuracy of the order of one per thousand. Theoretical calculations predict a lifetime that is in significant disagreement with this high-precision experimental value. Our mass shift calculations, based on a fully relativistic formulation of the nuclear recoil operator, are in excellent agreement with the experimental results and confirm the absolute necessity to include relativistic recoil corrections when evaluating mass shift contributions even in medium- Z ions.

1. Introduction

Measurements of transition energies, radiative transition probabilities and isotope shifts in few-electron systems represent a continuing challenge for theory because of the interplay of relativistic, correlation, quantum electrodynamic (QED), and nuclear recoil effects. Especially in the range of medium nuclear charges, these effects are all intertwined. Measurements of excited state lifetimes are particularly useful to test atomic structure theories since they are especially sensitive to the long-scale structure of the electronic wave function. Experimental investigations of optical isotope shifts in the spectra of highly charged ions (HCI) have the advantage of an increased sensitivity to nuclear effects. The study of mass shifts in few-electron ions of medium nuclear charge, besides providing the possibility of testing electron correlation calculations, is an ideal tool to investigate relativistic contributions to the nuclear recoil operator.

Here, we present results of an accurate lifetime measurement of the $1s^2 2s^2 2p^2 P_{3/2}$ metastable level of B-like Ar^{13+} in comparison with our theoretical calculations. Furthermore, the isotope shifts of this transition in B-like Ar^{13+} and of the $M1$ ground state transition in Be-like Ar^{14+} have been measured and calculated with high precision.

2. Lifetime of the $1s^2 2s^2 2p^2 P_{3/2}$ level in Ar^{13+}

The measurement was performed at the electron beam ion trap (EBIT) of the Max-Planck-Institut für Kernphysik [1]. In this experiment, the electron beam was periodically switched on (500 ms) to excite ions with energies well below the ionization threshold and, then, switched off (200 ms) to detect the transition. The lifetime of the $^2P_{3/2}$ level in B-like Ar^{13+} was measured by monitoring its decay to the $^2P_{1/2}$ ground state through an M1 transition and by analyzing the decay curves. After correcting a number of parameters influencing the lifetime, the final value was determined. For further details see Ref. [2]. Our experimental result of $\tau = 9.573(4)(5)$ ms (stat)(syst) is in agreement with the latest measured value of 9.70(15) ms [5] and it is more than an order of magnitude more accurate than three previous experiments [3, 4, 5].

A comparison with theoretical and previous experimental results is shown in Fig. 1. The theoretical results shown are as follows: MCDF [6], MCBP [7], C-S [8], CI-DFS [9], SS'98 [10], RQDO [11] and MCDF^a [12], and the previous experimental value is from Ref. [5]. A more recent calculation using the configuration interaction Dirac-Fock-Sturmian method (CI-DFS, Ref. [9]) leads to a theoretical value of $\tau = 9.538$ ms. The decay rate depends on the third power of the transition frequency which has been accurately measured recently (Ref. [13], $\lambda = 441.2559(1)$ nm). In order to give a more complete representation of the theoretical data, theoretical values are 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 electron anomalous magnetic moment (EAMM), which leads to a decrease of the lifetime by a relative factor of $1 - 2\alpha/\pi$. Theoretical lifetime results appear to scatter around a lifetime of 9.53 ms, which is within a 3σ disagreement with our experimental value. Presently there is no explanation for this discrepancy.

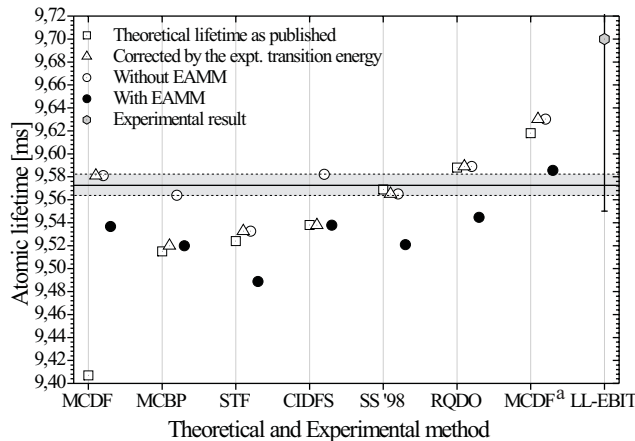


Figure 1. A comparison of the present result with calculated lifetimes of the $^2P_{3/2}$ state in Ar^{13+} [6, 7, 8, 9, 10, 11, 12] and (rightmost) the experimental result of Ref. [5]. The solid line indicates the mean value of the present data and the dotted lines represent its error. The electron anomalous magnetic moment (EAMM) contribution had been ignored in all theoretical calculations except for Refs. [7, 9]. (See the text and the references cited above for explanations.)

3. Relativistic many-body recoil effects in highly charged Ar ions

We investigated the isotope shift of the M1 transitions $1s^2 2s^2 2p^2 P_{1/2} - ^2P_{3/2}$ in Ar^{13+} and $1s^2 2s 2p^3 P_1 - ^3P_2$ in Ar^{14+} by performing accurate wavelength measurements with the isotopes ^{36}Ar and ^{40}Ar loaded into the EBIT. The corresponding wavelength shifts have been determined with sub-ppm accuracy. The details of the measurement can be found in Ref. [14].

Isotope shifts arise from the combined effect of the finite nuclear mass and volume on the electronic binding energy. The mass-dependent, or recoil part, is commonly divided into the so-called normal mass shift and the specific mass shift (mass polarization) contributions. The volume effect, also called the field shift (FS), is caused by the penetration of the electronic wave

function into the nuclear regime. The relativistic nuclear recoil operator for a system of atomic electrons is given by the formula (in relativistic units, see Refs. [15, 16])

$$R_{ij} = \frac{\mathbf{p}_i \cdot \mathbf{p}_j}{2M} - \frac{Z\alpha}{2Mr_i} \left(\boldsymbol{\alpha}_i + \frac{(\boldsymbol{\alpha}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^2} \right) \cdot \mathbf{p}_j. \quad (1)$$

Here, \mathbf{r}_i and \mathbf{p}_i are the position and the momentum vectors of the i th electron, respectively, and $\boldsymbol{\alpha}_i$ represents the Dirac matrices. Z is the atomic number, M is the mass of the nucleus and α denotes the fine-structure constant. The normal mass shift correction to a given atomic state is obtained as the expectation value of the diagonal sum of R_{ij} , whereas the specific mass shift term is given by the sum of the off-diagonal components $\langle \sum_{i \neq j} R_{ij} \rangle$. The first term in Eq. (1) corresponds to the mass shift operator also known in the nonrelativistic theory.

Transition energies and their respective isotope shifts, including the contributions due to the total relativistic recoil operator in Eq. (1) have been calculated by large-scale CI-DFS calculations [17]. A detailed breakdown of the normal and specific mass shift contributions NMS and SMS, together with the relativistic operator corrections RNMS and RSMS to the states under study is given in Table 1. The NMS and SMS nonrelativistic operator contributions were also calculated in the framework of the multiconfiguration DF method with independent coding [18] to provide a numerical test. The NMS, SMS, RNMS, and RSMS contributions in Table 1 define the nuclear recoil corrections within the $(\alpha Z)^4 m_e^2 / M$ approximation (m_e denotes the mass of the electron). The calculation of the QED contributions to the recoil effect is based on the works [19]. The calculation of these terms require using QED beyond the Breit approximation [15, 19]. QED corrections are given in the third and sixth rows of Table 1. The contributions due to the field shift (FS) effect are given in the seventh row of Table 1. In the case of the Ar isotopes studied here, the mass shift terms clearly dominate over the FS.

Table 1. Contributions of the relativistic recoil operator and QED recoil terms to the mass shift and field shift ($^{36}\text{Ar} - ^{40}\text{Ar}$) in Ar^{13+} and Ar^{14+} ions (in cm^{-1}). (See the text for explanations.)

	Ar^{13+}	Ar^{14+}
NMS	0.1052	0.0796
RNMS	-0.0822	-0.0627
One-electron QED	0.0002	0.0002
SMS	-0.0741	-0.0697
RSMS	0.1151	0.0887
Two-electron QED	-0.0008	-0.0015
FS	-0.0005	-0.0001
Sum	0.0629	0.0345

The final experimental and theoretical results for the transition wavelengths in Ar^{13+} and Ar^{14+} and their isotope shifts are presented in Table 2. Theoretical isotope shifts including relativistic normal and specific mass shift contributions show an excellent agreement with the measured results, confirming the relativistic theory of recoil effects in many-body systems [17]. A calculation with the nonrelativistic recoil operator would give values for the mass shift which are smaller than the correct relativistic result by approximately a factor of two (Ar^{13+}) or even three (Ar^{14+}), as it is evident from Table 1.

Table 2. Transition wavelengths for $^{40}\text{Ar}^{13+,14+}$ ions and isotope shifts ($^{36}\text{Ar} - ^{40}\text{Ar}$) (in nm, air). The π - and σ -components and their average (av) are given in the case of Ar^{13+} .

Ion	Wavelength (^{40}Ar)		Isotope shift ($^{36}\text{Ar} - ^{40}\text{Ar}$)	
	Theory	Experiment	Theory	Experiment
Ar^{13+}	441.16(27)	441.2556(1)	0.00123(5)	0.00120(10) $^{\pi}$
				0.00125(7) $^{\sigma}$
				0.00123(6) $^{\text{av}}$
Ar^{14+}	594.24(30)	594.3879(2)	0.00122(5)	0.00120(10)

4. Summary

The lifetime of the $\text{Ar}^{13+} 1s^2 2s^2 2p^2 P_{3/2}$ metastable state was experimentally determined to be 9.573(4)(5) ms (stat)(syst). The accuracy level of 0.1% makes this measurement sensitive to QED corrections like the electron anomalous magnetic moment and to relativistic correlation effects. Theoretical predictions, including recent calculations based on the configuration interaction Dirac–Fock–Sturmian method, cluster around a lifetime that is approximately 3σ shorter than the experimental result. At present we have no explanation for this interesting disagreement.

Isotope shifts of the ground state $1s^2 2s^2 2p^2 P_{3/2} - ^2P_{1/2}$ and $1s^2 2s 2p^3 P_1 - ^3P_2$ M1 lines in highly charged Ar^{13+} and Ar^{14+} ions, respectively, have been determined with high precision. The observed isotope effect has confirmed the relativistic theory of nuclear recoil effects in many-body systems, which removes major inconsistencies in earlier theoretical methods. A comparison with accurate calculations and a theoretical analysis of the contributing effects (see Table 1) shows that it is inevitable to take into account the *total* relativistic recoil operator when predicting relativistic mass shift contributions even in medium- Z ions. To our knowledge, the relativistic recoil effect has never been observed experimentally in HCI thus far.

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