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Isotope shifts of dielectronic resonances for heavy few-electron ions

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Abstract. Effects of the nuclear charge distribution upon the total cross-section of dielectronic recombination are investigated. We calculated isotope shifts of resonance energies for H-, He- and Li-like heavy ions in the relativistic domain from $Z = 54$ to $Z = 94$. We point out that the position of the resonances is most influenced in KLL transitions in very heavy elements and their shifts are most likely to be measurable for systems with intra-shell transitions. For these systems, dielectronic recombination experiments provide a new method to determine the parameters of the nuclear charge distribution.

PACS. 34.80.Lx Electron-ion recombination and electron attachment – 31.30.Jv Relativistic and quantum electrodynamic effects in atoms and molecules

1 Introduction

The dielectronic recombination (DR) is a resonance process. In the first step a free electron is captured radiationlessly by the simultaneous excitation of a bound electron, and in the second step a radiative transition occurs. The positions, strengths and widths of the resonances in few-electron high-Z ions deliver informations on the relativistic dynamics of electrons. It is known that the finite charge distribution of the nucleus leads to a remarkable shift of the resonance energies and has to be properly taken into account [1]. The preliminary calculations of Steih [1] gave rise to the hope to explore the nuclear charge distributions by measuring the isotope shifts of the resonances. The aim of this paper is now to study these isotope shifts as a function of the nuclear charge distribution in a systematic manner in order to assist the measurements of nuclear charge radii of exotic nuclei.

In Section 2 we outline the theory of DR and present the chosen details of the nuclear charge distribution. Section 3 contains the presentation and discussion of the numerical results on isotope shifts. We conclude with a short summary in Section 4.

Throughout this paper atomic units will be used: $e = m_e = \hbar = 4\pi\epsilon_0 = 1$.

2 Theory

In order to determine the radius of the nuclear charge distribution from the position of resonance lines observed in dielectronic recombination, we shortly review the formulas connected with this process.

2.1 Dielectronic recombination

The total cross-section for the dielectronic recombination process is given by using perturbative calculations and the projection operator method [2,3] as

$$\sigma^{DR}_{i\to f} = \sum_d \frac{2\pi^2}{\vec{p}^2}\frac{A_r}{\Gamma_d} L_d(E_{el} + E_i - E_d) V_a \tag{1}$$

with the Lorentz-like profile L_d of an intermediate state d at energy E_d

$$L_d(E_{el} + E_i - E_d) = \frac{\Gamma_d/2\pi}{(E_{el} + E_i - E_d)^2 + \Gamma_d^2/4}. \tag{2}$$

Here, E_{el} is the continuum energy of the captured electron and E_i the energy of the initial ion without the free electron. The free electron has an asymptotic momentum $\vec{p}$. The total width of the intermediate state, given as a sum of the radiative and Auger widths, is denoted by Γ_d. In deriving formula (1), the DR process was treated in the isolated resonance approximation and as independent from the radiative recombination (RR). These approximations have been proved to be of very good accuracy for heavy

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ions [4]. The further quantities in (1) are the resonant capture rate

$$V_a = \frac{2\pi}{2(2J_i+1)} \sum_{M_i m_s M_d} \int d\Omega_p \times |\langle \Psi_d; J_d M_d | V_{capt} | \Psi_i; J_i M_i, \vec{p} m_s \rangle|^2 \quad (3)$$

and the radiative decay rate from the intermediate state d to the final state f

$$A_r = \frac{2\pi}{2J_d+1} \sum_{M_f \lambda M_d} \int d\Omega_k \times |\langle \Psi_f; J_f M_f, \vec{k}, \lambda | H_{er} | \Psi_d; J_d M_d \rangle|^2 \rho_f. \quad (4)$$

The operator V_{capt}, responsible for the capture, is the sum of the Coulomb and Breit interactions. The initial state i of the system consists of bound electrons with a total angular momentum J_i and its projection M_i and the continuum electron with spin projection m_s. The resonant state d has the angular quantum numbers J_d and M_d. In (3) we average over the initial magnetic sub-states and the direction of the incoming electron, and sum over the quantum numbers of state d. The operator H_{er} in (4) describes the interaction between the electrons and the radiation field [5], and $\vec{k}$ and λ are the wave number vector and the polarization of the outgoing photon, respectively. The calculations of [6] show that the influence of a further photon emission on the total cross-section is negligible.

The energies of the bound states with approximate radiative corrections and the radiative decay rates were calculated using the multiconfigurational Dirac-Fock packages GRASP 1.0 [7] and GRASP92 [8]. Continuum orbitals and capture rates were generated by the AUGR module developed in [9] as an extension of the GRASP environment.

2.2 Isotope shifts and nuclear charge distribution

The position of the Lorentz peak in (2) depends on the energies of the initial and intermediate states, which are affected by isotopic effects. Isotope shifts are slight variations of the electron energies without splitting. For their calculation we assume a spherical charge distribution in form of a two-parameter Fermi distribution

$$\rho_{nuc}(r) = \frac{\rho_0}{1 + e^{(r-c)/a}}. \quad (5)$$

Here, c is the radius at which the density reaches half of its maximum value. The distance in the surface layer over which the density decreases from 90% to 10% of its maximum is

$$t = 4a \ln 3, \quad (6)$$

where t is referred to as the surface thickness of the distribution. The root mean square (RMS) radius is given by

$$r_{RMS} = \sqrt{\frac{1}{Z} \int_0^\infty r^2 \rho_{nuc}(r) 4\pi r^2 dr}. \quad (7)$$

We have chosen the distribution (5) because it is the most widespread one in the literature. Tabulations usually only list the RMS radius and the surface thickness parameter of the nucleus. Parameters of even more realistic models are only in single cases available. Compared with a point-like nucleus, the extended nuclear charge distribution leads to a shift of the resonance energies in the order of 200 eV in the case of the K shell in U, whereas the isotope shifts in U result in values of about 1 eV, as we demonstrate in the next section.

In most calculations, presented in Section 3, we used the following relation of Johnson and Soff [10] to obtain the RMS radius for a given mass number A:

$$r_{RMS} = \left(0.836\ A^{1/3} + 0.570\right) \text{fm} \quad (8)$$

and a fixed thickness parameter $t = 2.30$ fm. In all cases when experimental radii were available, we compared them with those delivered by formula (8). We found that the discrepancies of the isotope shifts of resonance energies are negligible on the requested level of accuracy of 10^{-2} eV.

In addition to the volume effect described above, small shifts arise due to the change of the nuclear mass in isotope series. In the nonrelativistic theory of nuclear motional effects, the mass shift is the sum of the normal mass shift and the specific mass shift [11]. The normal mass shift is taken into account by replacing the electron mass m_e with the reduced mass

$$\mu = \frac{m_e M_{nuc}}{m_e + M_{nuc}}, \quad (9)$$

where the nuclear mass M_{nuc} is isotope dependent. This replacement is only valid in the non-relativistic limit, but it is used as an approximation in the relativistic case. The factor μ/m_e used to multiply the energies calculated with the assumption of a nucleus with infinite mass is approximately $1-10^{-6}$ for all elements considered in the present paper, i.e. the reduced mass shift contribution is 6 orders of magnitude smaller than the volume shift.

As calculations performed with the GRASP92 package [8] revealed, the specific mass shift is also negligible. The negligibility of the nuclear mass shift allows one to study the volume shift effect in an unperturbed way.

3 Numerical results

In this section we present numerical results for total DR cross-sections and resonance energies under the influence of the charge extension of the nucleus. The calculations were performed for relativistic ions in the range from $Z = 54$ to $Z = 94$. We have investigated isotope shift effects in three different ionic systems, namely, for H-, He- and Li-like ions.

For Pb isotopes, Figure 1 shows the typical behavior of the total DR cross-section when the nuclear charge distribution changes with the mass number. The curves in the figure present the resonance $[1s_{1/2}(2p_{3/2}^2)_2]_{5/2}$ for capture

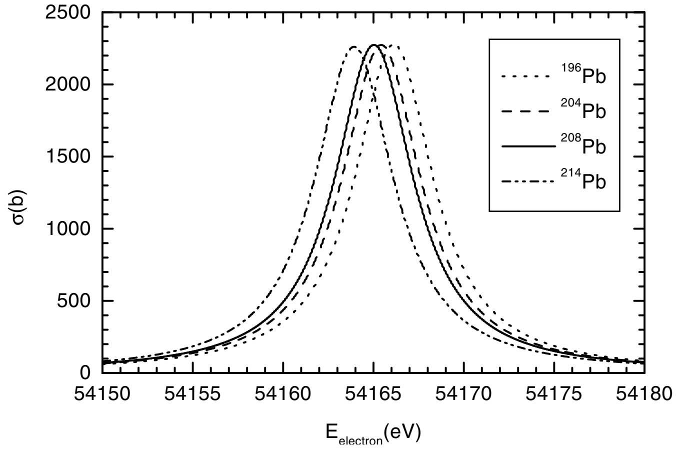

Fig. 1. Total DR cross-section for He-like Pb, at the energy of the $[1s_{1/2}(2p^2_{3/2})_2]_{5/2}$ resonance, as a function of the continuum electron energy.

into He-like Pb ions. Only in this calculation, we used nuclear RMS radii and the thickness parameters from the comprehensive compilation of Fricke et al. [13]. The figure shows that the energy position of the peak is shifted to the right when a lighter isotope, i.e. a smaller RMS radius, is considered. However, the shape of the cross-section, i.e. the maximum value and the width of the peak, does not change.

Similar calculations were performed for a wide range of elements and electron energies in H-, He-, Li-like systems. In all cases, the characteristics of the resonance shapes in the cross-sections are not noticeably affected. This allows us to restrict ourselves in the following to the shift of resonance energies.

In Figures 2–5 we demonstrate the dependence of isotope shift of resonance energies on the charge number Z for some selected resonances. The shift is calculated as the difference between the resonance energies for a given mass number A and for $A-5$, respectively,

$$\Delta E_{res}(Z,A) = E_{res}(Z,A-5) - E_{res}(Z,A). \qquad (10)$$

The mass number A, corresponding to a stable isotope, is obtained by inverting the semi-empirical formula

$$Z = \frac{A}{1.98 + 0.015A^{2/3}} \qquad (11)$$

and rounding the mass number A to an integer value. Expression (11) can be derived from the Weizsäcker mass formula of the nuclear drop model [14]. The RMS radius and the surface thickness were taken from (8).

Figure 2 shows the Z-dependence of the isotopic energy shifts of 5 selected resonances out of 10 possible ones for KLL DR into H-like heavy ions. In ions with higher charge numbers, the electronic probability density has a larger overlap with the nuclear matter because of the strong electrostatic attraction. This results in more pronounced nuclear volume effects of the energy levels. By fitting a power function on the curves of Figure 2 we found that these resonance energy shifts scale as $Z^{6.2} \ldots Z^{6.5}$.

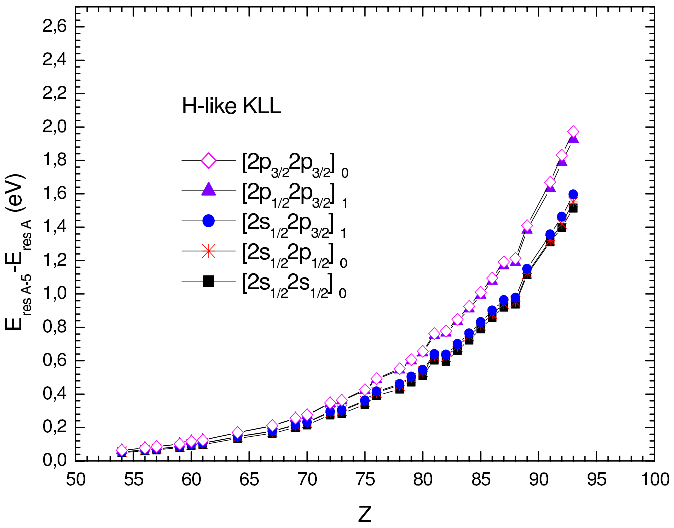

Fig. 2. Difference between the resonance energies of two different isotopes with mass numbers A and $A-5$ in the case of KLL DR into H-like ions as a function of the charge number.

The resonances with both electrons in the $2p_{3/2}$ state are shifted most. This can be understood if one keeps in mind that the resonance energy is the difference of the initial and intermediate state energies, so for its isotope shift $\Delta E_{res} = \Delta E_i - \Delta E_d$ holds. In the case of KLL DR, i is a $1s$ state, which is far more sensitive to nuclear properties than any other one-electron state. As a consequence, the resonance energy shift originates dominantly from the shift of the initial state. Because of the subtraction in ΔE_{res}, the shift of the intermediate state counteracts the initial state shift. Knowing that within the L-shell the $2s$ wave function has the largest overlap with the nucleus and the $2p_{3/2}$ function the least, the ordering of the curves in Figure 2 can be well understood.

As already mentioned before, we evaluate the quantum electrodynamic corrections to the energy levels by means of the methods used in the GRASP packages. Using the results of the more elaborate calculations of Beier et al. [15] for the RMS radius dependence of the QED corrections of order α in H-like Th ($Z = 90$), we can conservatively estimate the error of the isotopic shift of resonance energies due to the approximative inclusion of radiative effects to be in the range of 10^{-2} eV, even in the case of elements as heavy as Th. This is the main source of errors in our computations.

We proved with separate calculations that the correlation energy in many-electron systems is weakly influenced by nuclear properties. Using different correlation configurations and different self-consistent schemes (the Optimal Level and Avarage Level methods, see [7]), we reach the conclusion that the level of sophistication in the self-consistent procedures only affects isotopic resonance energy shifts below the meV level. This is due to the fact that the part of the electronic wavefunction inside the nucleus is weakly influenced by the other electrons in the shell. The effect of the Breit interaction was also considered as described in [8]. Similarly, its influence on the isotopic shift

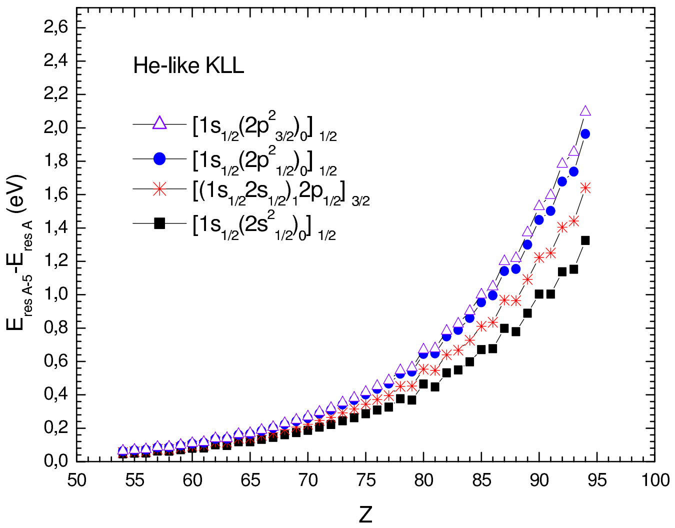

Fig. 3. Difference between the resonance energies of two different isotopes with mass numbers A and $A-5$ in the case of KLL DR into He-like ions as a function of the charge number.

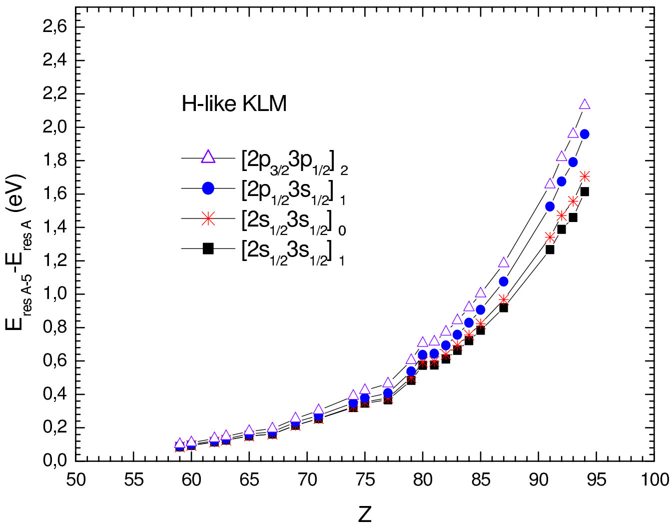

Fig. 4. Difference between the resonance energies of two different isotopes with mass numbers A and $A-5$ in the case of KLM DR into H-like ions as a function of the charge number.

of resonance energies is in the range of meV. Nevertheless, the energy shifts were all calculated by means of the most sophisticated method of those mentioned above. The only observable difference between self-consistent calculations and those not including the electron interaction at all occurs in our study of the $1s^2$ and $2s^2$ states. In all other cases, the isotopic energy shift of the two-electron states is just the sum of the corresponding one-electron state shifts. As a result, the shift of the intermediate state is also independent of the coupling of the orbital wave functions. For instance, the volume shift of the $[2p_{3/2}2p_{3/2}]_2$ resonance is the same as that of the $[2p_{3/2}2p_{3/2}]_0$ resonance. Another advantage is that even if the absolute energy position of a peak is calculated with an error of a few eV, its isotopic shift is determined much more accurately. An error estimate is presently not possible.

The same analysis can be repeated for KLL recombination into He-like ions given in Figure 3. In this case, there is an extra electron in the K-shell. Its presence modifies the total energy of the states involved in the process. Since in most cases the correlation energy is independent of the nuclear properties, the curves in Figures 2 and 3 look very similar. From the point of view of an experimental implementation, the charge state of the ion is not of great importance when one wants to measure nuclear volume shifts. The same is expected for Li- and Be-like ions, as long as KLL recombination is considered.

Figure 4 presents the isotope shift of resonance energies in the case of KLM DR into H-like ions. In this process, the $n=3$ states are also involved. They are slightly less sensitive to the charge distribution of the nucleus than the L-states, so the resonance energy shifts are a bit larger than for KLL resonances. However, these differences are not significant, hence it is sufficient to perform measurements at the lower KLL energies. We repeated these KLM calculations for capture into He-like ions. The results are basically equivalent to those given in Figure 4. This again proves that the electron correlation effects are independent of the nuclear charge distribution.

The resonances of the KLL DR in heavy systems have energies of the order of 50 keV. In this regime the measurement of the resonance energies is presently not sufficiently precise to detect isotope shifts. In contrast to this, it is possible to measure the energies of the resonances of dielectronic capture in Li-like ions with a quite high precision since the energy of the free electron is of the order of few eV [16]. A capture with a transition within the L-shell is e.g.

$$A^{(Z-3)+}(1s^2 2s) + e^- \to A^{(Z-4)+**}(1s^2 2p_{3/2} 6l_j) \to A^{(Z-4)+*} + \gamma. \quad (12)$$

For this DR we have calculated the isotope shifts as a function of Z as shown in Figure 5. The shifts are much smaller than in the previous cases. The electrons in the $n=2$ and $n=6$ shells are both far away from the nucleus, therefore, the initial and intermediate states of the DR process are almost equally shifted by a change of the mass number of the element. However, shifts in the range of 0.1 eV are presently measurable [16].

4 Summary

In this paper we studied the influence of the nuclear charge distribution on the position of the resonances in the cross-section of dielectronic recombination. We performed fully relativistic calculations with the multiconfigurational Dirac-Fock program GRASP to determine energies, widths and resonance strengths of DR peaks. Different reaction pathways with capture into H-, He- and Li-like ions were considered for elements in the range between $Z=54$ and $Z=94$. We found that isotopic variations in the nuclear charge distribution parameters result in shifts of the resonance energies, but the resonance

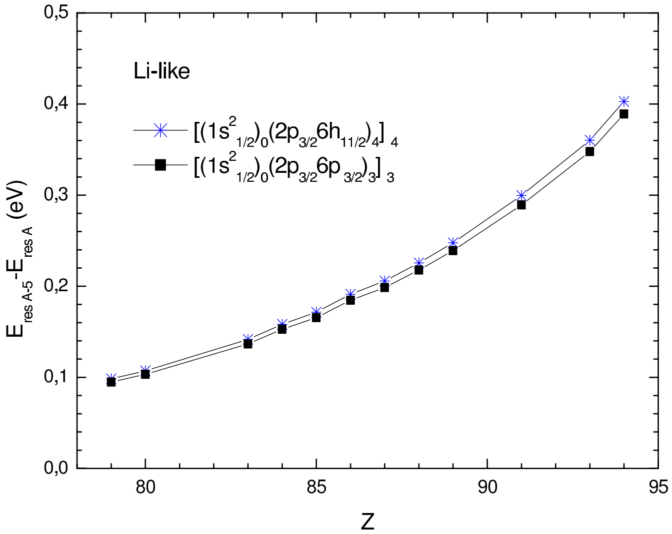

Fig. 5. Difference between the resonance energies of two different isotopes with mass numbers A and $A-5$ in Li-like ions as a function of the charge number.

shapes remain practically unchanged. Further, we analyzed the dependence of resonance energy shifts on the charge number Z, on the set of intermediate states as well as on the number of electrons involved in the recombination process. Our results give the largest isotope shifts for a recombination with very heavy ions where a K-shell electron is excited, and the largest relative shifts $\Delta E_{res}/E_{res}$ for low-lying resonances related to intra-shell transitions. Therefore, these systems are most appropriate in future for an experimental investigation of nuclear charge radii. The number of electrons present in the ion and their correlations are of minor importance.

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