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Similarity between the angular distributions of the first- and second-step electrons in sequential two-photon atomic double ionization

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ABSTRACT

Angular distributions of electrons produced in sequential two-photon atomic double ionization are considered theoretically by comparing the results obtained for the first and the second electron. It is shown analytically that the angular distributions of the two emitted electrons in many cases are similar for ionization from a closed-shell atom. Numerical examples of this counter-intuitive similarity are presented. In addition, examples are given demonstrating the difference between the angular distributions of the first photoelectron in sequential two-photon double ionization and in conventional one-photon single ionization.

1. Introduction

Angle-resolved photoelectron spectroscopy is a powerful tool for the study of small quantum systems, such as atoms, molecules, and nanostructures. Angular distributions and angular correlations of the emitted electrons provide valuable information on the electronic structure of these systems and the dynamics of their interaction with photons. In spite of this, photoelectron spectroscopy of ions received a limited attention because of difficulties in producing the ionic target with sufficient density [1–3]. The corresponding experiments became much more feasible only recently with the advent of the XUV and X-ray free electron lasers (FELs) with their unprecedented intensity in this range of the photon energies, concentrated in short femtosecond pulses. Photoelectron spectroscopy of the sequential two-photon double ionization (2PDI) is one of the directions that began to be actively developed with the FELs. Moreover, the 2PDI was among the first phenomena observed with the first FEL operation in the XUV wavelength range, FLASH in Hamburg [4].

In accordance with the current stage of the studies, we exemplify here this process by the sequential 2PDI of the outer shell of noble gas atoms, which when treated as a stepwise process can be written as follows (see Fig. 1(a)):

Step 1: Photoionization of a neutral atom with formation of the singly charged ion:

$$\gamma + \mathrm{A}(\mathrm{np}^6\,{}^1\mathrm{S}_0) \rightarrow \mathrm{A}^+(\mathrm{np}^5\,{}^2\mathrm{P}_{\frac{1}{2},\frac{3}{2}}) + \mathrm{e}_1. \quad (1)$$

Step 2: Further ionization of the singly charged ion by a photon from the same XFEL pulse with formation of the doubly charged ion:

$$\gamma + \mathrm{A}^+(\mathrm{np}^5\,{}^2\mathrm{P}_{\frac{1}{2},\frac{3}{2}}) \rightarrow \mathrm{A}^{2+}(np^4\,{}^3\mathrm{P}_{0,1,2},\ {}^1\mathrm{D}_2,\ {}^1\mathrm{S}_0) + \mathrm{e}_2. \quad (2)$$

The photoelectron lines corresponding to the emission of electrons e_1 and e_2 are well separated in energy due to the different ionization thresholds of positive ions and neutral atoms (Fig. 1(b)). Strong observed signals from the sequential 2PDI are caused by the large intensity and the short pulse duration of the FEL. Indeed, despite of the Coulomb repulsion in the ionic target, the singly charged ions are heavy enough to keep their positions and therefore to maintain a high ionic density within a few femtoseconds between the first and the second ionization steps. The sequential 2PDI dominates over the direct two-photon double ionization when the photon

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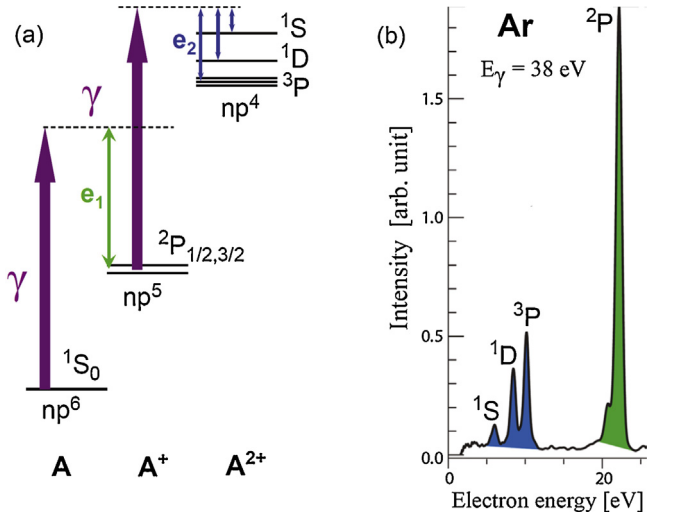

Fig. 1. (a) Scheme of the 2PDI in a noble gas atom and (b) the corresponding photoelectron spectrum for the specific case of atomic argon measured at the photon energy of 38 eV. Panel (b) after Braune et al. [4].

energy exceeds the ionization threshold of the single charged ion [5–7]. In case of direct double ionization, the electrons e_1 and e_2 share the excess energy in a smooth way, and the corresponding photoelectron spectrum does no longer contain the characteristic lines.

The photoelectron lines from the second step (2) are strong enough (see Fig. 1(b)) that also the photoelectron angular distribution (PAD) of the electrons e_2 can be measured, as it has been demonstrated in a few experiments on noble gas atoms [4,8–11]. Moreover, the angular correlations between the electrons e_1 and e_2 have been observed in experiments on the neon atom at FLASH, although with low statistics [12]. The high intensity of the photoelectron lines in the ionization of the ions allows, in principle, to think on realizing a set of experiments similar to the case of neutral atoms, including spin-sensitive measurements and performing a complete experiment on the ionic photoionization [13].

Theoretical developments, performed shortly after the first measurements on the PADs in sequential 2PDI, rather successfully explained the experimental results on the photoelectron spectra and PADs of the electrons e_2 [14,15]. Also, the PADs of the third-step electrons in the sequential three-photon triple ionization have been observed at FLASH and successfully described theoretically [16]. Furthermore, the PADs of the second-step electrons in the three-photon double ionization [17,18], observed at SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [19,17,20] have been studied, as well as the already mentioned angular correlations in the sequential 2PDI of neon [12,21,22]. A comprehensive review of these activities is contained in [23].

Against the backdrop of the above advances it is remarkable that the PAD of the first-step electron e_1 has not received much attention in the experiments on the sequential 2PDI, despite its predicted unusual shape. We stress once more that speaking about the electrons e_1, only double ionization is implied. In this context, for example, the line ^2P in Fig. 1(b) does not represent solely the electrons e_1: this line, measured irrespective to the final argon ion charge, is a sum of contributions from electrons emitted in single and double (or even multiple) ionization in an unknown proportion. It was shown theoretically [15] that the PAD of the electron e_1 in the dipole approximation deviates from the standard distribution of the form

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{4\pi}\left(1 + \beta P_2(\cos\theta)\right), \tag{3}$$

where a linearly polarized FEL beam is implied, the angle θ is counted from the direction of this linear polarization, $P_k(x)$ is the Legendre polynomial, β is the asymmetry parameter, and σ_0 is the angle-integrated cross section. When the second-step ionization occurs, the PAD (3) for electrons e_1 generally transforms into

$$\frac{d\sigma}{d\Omega} = \frac{\sigma_0}{4\pi}\left(1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)\right), \tag{4}$$

with an additional third term. The form (4) applies even if the second electron remains undetected: it is enough that the second electron is emitted, albeit its direction of emission and its energy are not determined. It gives the impression that the electron e_1 'knows' whether the second electron e_2 is emitted or not: the PAD of e_1 is different if it is measured in coincidence with the singly or doubly charged ions as produced in the photoionization process. This interesting phenomenon has been discussed in [15,24,23]. The probability of the 2PDI is the product of the PAD (3) and the total probability of the second-step photoionization. The latter depends on the emission angle of the first electron, since the intermediate state is aligned and the alignment depends on this angle. This makes the angular distribution of the first electron more complex. We stress here that with the conventional synchrotron radiation sources, the probability of the sequential 2PDI is negligible in comparison with the single-photon ionization and therefore (within the dipole approximation) the contribution of the third term into the PAD (4) was even not mentioned. Only with the recent advent of FELs, the probability for producing doubly charged ions has become comparable to that for single photoionization, and modifications to the first-step PAD beyond the standard form (3) have attracted attention.

Noteworthy, the PAD of the electron e_2, which is of the form (4), is naturally explained by the fact that the intermediate ion A^+ is aligned after absorption of the first (linearly polarized) photon, causing the nonvanishing β_4, which is proportional to the alignment [15].

In the context of comparing the angular distributions of the electrons e_1 and e_2 in stepwise processes, it is instructive to discuss a similar problem in another two-step reaction such as the photoinduced Auger decay:

$$\gamma + A \longrightarrow A^{+*} + e_1 \tag{5}$$

$$\hookrightarrow A^{2+} + e_2\,, \tag{6}$$

where e_1 and e_2 are the photoelectron and the Auger electron, respectively. The process (5)+(6) differs from the process (1)+(2) in the sense that the second-step ionization is caused by the Auger decay and not by photon absorption. In the case of the photoinduced Auger decay (5)+(6), integrating the angular correlation function of the electrons e_1 and e_2 over the emission angles of the electron e_2 results in the conventional PAD of the electron e_1 of the form (3) (for an unpolarized atom A). This is in contrast to the sequential 2PDI, where the similar procedure gives the PAD of electron e_1 in the form (4). The reason for this lies in the fact that the absorption of the second photon, which always possesses a distinguished direction (the direction of the photon propagation or its polarization) in (2), is anisotropic by its nature, while the Auger decay in (6) is governed by the rotationally invariant scalar interaction.

However, turning to the sequential 2PDI, our numerical calculations predict that the influence of the second-step photoionization on the PAD of the first-step electrons smears out to a large extent after their PADs are summed over the final ionic multiplet states of $A^{2+}(np^4)$, i.e. over the energies of the unobserved electron e_2. Therefore to observe the nontrivial PAD of the electron e_1 in the sequential 2PDI, it is preferential to measure either the state of the final doubly charged ion or the kinetic energy of the

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