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CALCULATIONS OF PHOTOELECTRON EMISSION SPECTRA RESULTING FROM ATOMIC $1s$ SHELL PHOTOIONIZATION FOLLOWED BY SEQUENTIAL RADIATIVE AND AUGER DECAYS OF INNER VACANCIES

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Abstract. Photoelectron emission from $1s$ shell photoionization of an argon atom and a carbonyl sulfide (OCS) molecule followed by sequential X-ray radiative and Auger decays has been theoretically studied within *ab initio* quantum many-body approach. Energy spectra of electron emission are calculated for photoelectrons registered in non-coincident with Auger electrons measurements and compared with experimental data. Prominent postcollision interaction distortion of the photoelectron energy spectra is reported. The quantum many-body approach employed provides more accurate description of photoelectron emission than simple approximate model used earlier.

Keywords: inner-shell photoionization, postcollision interaction, Auger decay, argon atom, carbonyl sulfide molecule

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РАСЧЕТ СПЕКТРОВ ФОТОЭЛЕКТРОННОЙ ЭМИССИИ, ВОЗНИКАЮЩЕЙ ПРИ ФОТОИОНИЗАЦИИ АТОМНЫХ $1s$ -ОБОЛОЧЕК, СОПРОВОЖДАЮЩЕЙСЯ ПОСЛЕДУЮЩИМИ РАДИАЦИОННЫМ И ОЖЕ РАСПАДАМИ ВНУТРЕННИХ ВАКАНСИЙ

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Аннотация. В работе исследована методом квантовой теории многих тел фотоионизация $1s$ -оболочки атома аргона и молекулы сульфида карбонила (OCS) с последующими радиационным и Оже распадами внутренних вакансий. Энергетические спектры электронной эмиссии были рассчитаны для электронов, регистрируемых без совпадения с

Оже-электронами, и сравнены с ранее полученными экспериментальными спектрами. Обнаружено заметное искажение спектров фотоэлектронов, вызванное послестолкновительным взаимодействием. Примененный метод квантовой теории многих тел обеспечил лучшее согласие с экспериментом, по сравнению с простой приближенной моделью, используемой ранее.

Ключевые слова: фотоионизация глубоких оболочек, послестолкновительное взаимодействие, Оже-распад, атом аргона, сульфид карбонила

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Introduction

Photoionization of a deep atomic shell leads to the formation of an inner vacancy. The subsequent decay of this vacancy can occur through various channels including radiative and non-radiative processes. The most common example of non-radiative decay channel is the Auger decay of an inner vacancy with the emission of a fast Auger electron. This process called the single Auger decay (SA) is well known and has been widely studied in recent several decades. An interest of researchers is related to the fact that three resulting particles, namely, a photoelectron, an Auger electron, and a residual ion interact with each other by the Coulomb forces. Such interaction leads to a strong distortion of the spectra of emitted electrons: these spectra are shifted and their line shapes become asymmetrical. This effect is known as postcollision interaction (PCI) and has been well studied both experimentally [1 – 5] and theoretically within a number of models [6 – 13].

Another way of the deep vacancy decay can occur via the radiative transition. This process is becoming increasingly important for K -shell vacancies of heavy atoms since the fluorescence yields increase rapidly with the atomic number Z [14, 15]. In this case, an electron of higher inner shell fills in the deep vacancy with creation of the intermediate inner-shell vacancy and emission of a photon. After that, in the next step, the intermediate inner-shell vacancy undergoes Auger decay with the fast Auger electron emission. Such a process of the sequential radiative and Auger decays (SRAD) of an inner vacancy has been studied mainly experimentally in the last decade [16 – 23]. Both coincidence and noncoincidence measurements of the photoelectron spectra and the Auger electron spectra reveal the PCI distortion of the measured electron spectra.

The proper description of PCI effects in the SRAD processes should take into account the energy sharing between three emitted particles, namely, a photoelectron, an Auger electron and an emitted photon. Instead, the first attempts of theoretical consideration of the SRAD process reduced the problem to a simpler SA decay with some effective decay width of the deep vacancy [17]. Within this model, the real three-step SRAD process is replaced by the two-step photoionization process of the deep shell followed by the SA decay of created vacancy where the real lifetime width of this vacancy is changed to some effective value Γ_{eff} . The Γ_{eff} value is chosen to adjust the decay time of an inner vacancy to cumulative time of sequential radiative decay, $\tau_r = 1/\Gamma_{1s}$, and the Auger decay, $\tau_A = 1/\Gamma_{2p}$, namely, $\Gamma_{eff} = 1/(\tau_r + \tau_A)$.

The more sophisticated approach for the description of the Auger electron spectra in the SRAD process developed in Ref. [21] on the basis of the model of the multiple Auger decay (proposed earlier in Ref. [24]) considers the SRAD as a three-step process. However, this approach as well as the simpler effective width model does not take into account the participation of the emitted photon in the energy sharing. Both the model of the effective width (see Ref. [17]) and the quantum model of Ref. [21] show a good agreement with noncoincidence measurements of the PCI distorted Auger electron spectra [21, 22]. However, an application of the effective width model to the noncoincidence measurements of the photoelectron spectra shows less satisfactory



agreement [17]. This disagreement reflects a serious shortcoming of the approximations [17, 21], namely, these models do not take into account the participation of the emitted photon in the PCI energy exchange.

Meanwhile, an *ab initio* quantum many-body approach for the SRAD process was recently developed in Ref. [16]. Calculations of the PCI distortion of the electron spectra within this approach showed an excellent agreement with both the noncoincidence measurements of the Auger electron spectra and the photoelectron spectra measured in coincidence with the Auger electrons [16]. However, a comparison of the available experimental data on the noncoincident photoelectron spectra [17, 25] with the results of the developed approach [16] has not been done so far. In experimental studies [17, 25] the Auger electrons were not registered while the photoelectrons were detected in coincidence with the ion valence. In Ref. [17], the spectra of photoelectron emission were studied in the SRAD process of the argon 1s shell photoionization. The similar process of the 1s shell photoionization of the S atom in the OCS molecule has been studied in Ref. [25].

In the present paper, we have applied our quantum many-body theory [16] to the SRAD photoionization processes studied experimentally in Refs. [17, 25]. We have compared the experimental spectra with results of *ab initio* quantum many-body approach and the effective width model. A comparison has revealed a noticeable difference between the experimental results and predictions of the effective width model and has showed rather well agreement with calculations results of our quantum many-body approach.

Our paper is organized as follows: in the next section we outline shortly our theoretical approach [16]. In the section following it, we present the calculated photoelectron spectra and compare them with experimental results [17, 25]. The atomic system of units, $|e| = m_e = \hbar = 1$, is used throughout the paper.

Photoelectron spectra in the SRAD process

An emission of a photoelectron from the deep 1s atomic shell followed by the SRAD decay of inner vacancy is a three-step cascade process which can be presented by the following scheme:

$$\gamma_0 + A \rightarrow A^{+*}(1s^{-1}) + e_{ph} \rightarrow A^{+*}(2p^{-1}) + \gamma_1 + e_{ph} \rightarrow A_f^{2+}(3p^{-2}) + \gamma_1 + e_{ph} + e_A. \quad (1)$$

The first step is the ionization of the 1s shell of the atomic target A by the incident photon γ_0 of energy ω_0 resulting in emission of a photoelectron e_{ph} and creation of a quasi-stationary $1s^{-1}$ vacancy. The second step is the radiative decay of the $1s^{-1}$ vacancy resulting in emission of a photon γ_1 of energy ω_1 and creation of an intermediate quasi-stationary $2p^{-1}$ vacancy. The last step is an Auger decay of the $2p^{-1}$ vacancy resulting in emission of an Auger electron e_A of energy E_A leaving the residual ion with two vacancies in the outer shell, $A_f^{2+}(3p^{-2})$.

An amplitude of the process (1) was recently developed in Ref. [16] by means of the quantum many-body theory:

$$A_{SRAD} = \frac{iM_0 M_1 M_A \left(\langle \chi_{E_{ph}} | \chi_{\omega_0 + E_{1s} + \frac{i}{2}\Gamma_1}^{(+)} \rangle - \langle \chi_{E_{ph}} | \chi_{\omega_0 - \omega_1 + E_{2p} + \frac{i}{2}\Gamma_2}^{(+)} \rangle \right)}{\omega_1 - E_{2p} + E_{1s} - \frac{i}{2}(\Gamma_2 - \Gamma_1)}, \quad (2)$$

where M_0 , M_1 , M_A are the matrix elements of photoabsorption, photoemission and Auger decay, respectively, that are weakly dependent on the energies of the emitted electrons; E_{1s} , E_{2p} are the 1s shell and 2p shell electron energies, respectively; $\Gamma_{1,2}$ denotes the decay widths of $1s^{-1}$ and $2p^{-1}$ vacancies, respectively; $\langle \chi | \chi^{(+)} \rangle$ are the overlap integrals between the photoelectron radial wave functions in the intermediate and final states before and after the Auger decay.

The wave function of the final electron state $\chi_{E_{ph}}$ is a standing wave which describes the motion of the outgoing photoelectron with energy E_{ph} in the Coulomb fields of the double charged residual ion and the Auger electron. The wave function of an intermediate electron state $\chi_E^{(+)}$ behaves asymptotically as an outgoing wave and describes the photoelectron motion in the field of a single charged ion before the Auger decay. Radiative decay of the $1s^{-1}$ vacancy does not change the charge of the ionized atom A^{+*} . Therefore, it does not alternate the long-range Coulomb field acting on the outgoing photoelectron. Indeed, its typical distance from the target

atom, at the moment of the $1s^{-1}$ vacancy decay, is

$$r \sim v_{ph} \tau_r = v_{ph} / \Gamma_1 \gg 1.$$

This crucial fact that the PCI takes place at large distances is related with small values of the inner vacancies decay width, $\Gamma \ll 1$.

There are two overlap integrals in the right-hand side of Eq. (2). In the first one, the energy of intermediate electron state corresponds to the stage after the ionization and prior the radiative decay:

$$E_1 = \omega_0 + E_{1s} + (i/2) \Gamma_1 = \Delta E + (i/2) \Gamma_1. \quad (3)$$

This energy is complex because the ionized atom $A^{+*}(1s^{-1})$ with deep $1s^{-1}$ vacancy is metastable. Its imaginary part corresponds to the $1s^{-1}$ vacancy decay width Γ_1 , and its real part

$$\text{Re}\{E_1\} = \omega_0 + E_{1s} = \Delta E$$

is the excess photon energy above the $1s$ shell ionization threshold.

Consequently, the amplitude of the intermediate photoelectron wave function slowly decays as $|\chi_E^{(+)}|^2 \propto \exp(-\Gamma_1 r / v_{ph})$ which confirms the estimation of typical PCI distances made above.

The energy of a photoelectron intermediate state in the second overlap integral

$$E_2 = \omega_0 - \omega_1 + E_{2p} + (i/2) \Gamma_2 = \Delta E - \omega_1 + E_{2p} - E_{1s} + (i/2) \Gamma_2 = \Delta E - \delta\omega_1 + (i/2) \Gamma_2 \quad (4)$$

corresponds to the stage between the radiative decay of $1s^{-1}$ vacancy with emission of a photon γ_1 of energy ω_1 and creation of the intermediate $2p^{-1}$ vacancy and the Auger decay of the $2p^{-1}$ vacancy. Its real part differs from the excess photon energy ΔE by the energy distance of the emission photon energy from its resonance value

$$\delta\omega_1 = \omega_1 - E_{2p} + E_{1s},$$

and its imaginary part corresponds to the $2p^{-1}$ vacancy decay width Γ_2 .

The square of modulus of the amplitude $A_{\text{SRAD}}(\omega_0, E_{ph}, E_A)$ determines the triple differential cross section of the process (1):

$$\frac{d^3\sigma}{dE_{ph}d\omega_1dE_A} \propto |A_{\text{SRAD}}(\Delta E, E_{ph}, E_A)|^2 \delta(E_{ph} + E_A + \omega_1 - 2E_{3p} - \omega_0). \quad (5)$$

It is worth to note that the amplitude A_{SRAD} (see Eq. (2)) depends on the incident photon energy ω_0 and the energies of three emitted particles ω_1, E_{ph}, E_A . But, due to the energy conservation law

$$E_{ph} + E_A + \omega_1 - 2E_{3p} - \omega_0 = 0, \quad (6)$$

two final energies only, e.g., E_{ph} and E_A , can be considered as independent parameters, as it is written by Eq. (5).

In this paper, we have analyzed the spectra of the photoelectrons measured in the SRAD processes in coincidence with the valence of the residual ion only. It means that the energy E_A of the Auger electrons as well as the emitted photon energy ω_1 is not fixed and a yield of the Auger electrons with all possible energies contributes to the measured photoelectron spectrum. Hence, the measured photoelectron yield is given by the triple differential cross section (see Eq. (5)) integrated over all the Auger electron E_A and photon ω_1 energies:

$$\frac{d\sigma}{dE_{ph}} = \int_0^\infty |A_{\text{SRAD}}(\omega_0, E_{ph}, E_A)|^2 dE_A. \quad (7)$$

Results and discussion

The energy spectra of photoelectrons emitted in the $1s$ shell ionization followed by the SRAD decay of an inner vacancy were earlier measured for the Ar atom [17] and for the S atom in the OCS molecule [25]. The SRAD process is the main channel of the $1s^{-1}$ vacancy decay of the systems considered which leads to the double charged residual ion [15, 26, 27]. Hence, the coincidences of photoelectron emission with the ion valence used in the measurements [17, 25] can guarantee that the main contribution to the measured photoelectron spectra comes from the process (1).

We have calculated the photoelectron spectra of the Ar atom and S atom according to the Eqs. (2) – (7). The overlap integrals in the amplitude $A_{\text{SRAD}}(\omega_0, E_{ph}, E_f)$ were evaluated within the semiclassical Wentzel – Kramers – Brillouin (WKB) approximation [12, 16]. This approximation is widely used for such calculations because the PCI takes place at large distances from the target atom where the electronic motion is semiclassical. The main advantage of WKB approximation is that it gives the closed analytical expression for overlap integral (see e.g., Refs. [12, 16]).

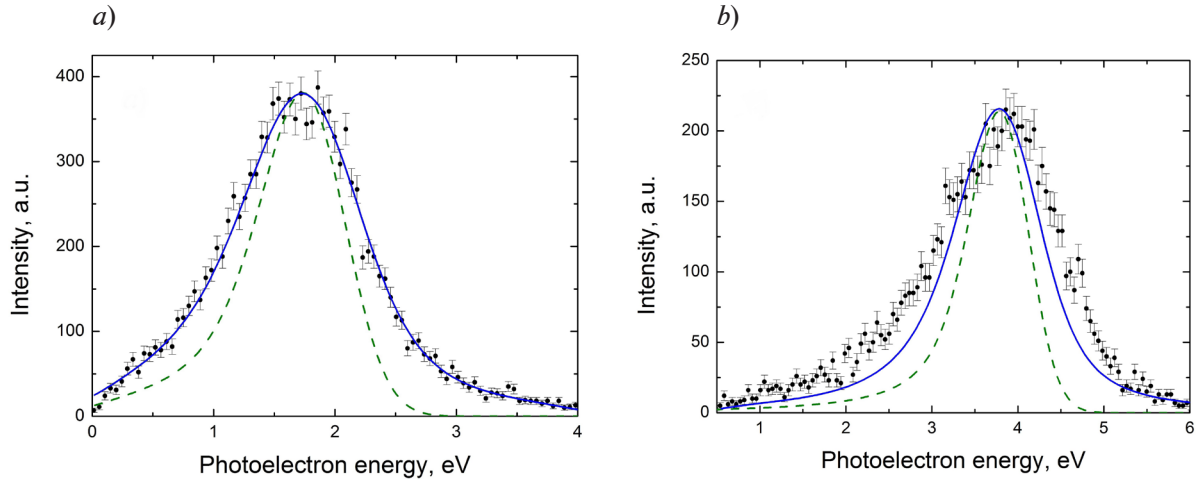


Fig. 1. Experimental (black circles) [17] and calculated (blue solid lines) photoelectron energy spectra in Ar $1s^2$ photoionization followed by the SRAD process for the excess photon energy $\Delta E = 2.0$ eV (a) and $\Delta E = 4.0$ eV (b). The calculations have been carried out within the quantum many-body approach according to Eqs. (2) – (7); the green dashed lines present the results of the effective width model

For Ar atom the parameters of the SRAD process are as follows [16]:

the $1s$ photoionization threshold energy $E_{1s} = -3206.3$ eV [28],

the energy of radiative transition $E_{2p} - E_{1s} = 2957$ eV,

the decay width of the vacancy is $\Gamma_1 = 0.69$ eV [14],

for the $2p$ vacancy the decay width is $\Gamma_2 = 0.118$ eV [29].

For the S atom in the OCS molecule we have used the following values of the widths:

$\Gamma_{1s} = 600$ meV and $\Gamma_{2p} = 60$ meV [14].

Finally, the cross section (7) was convoluted with a Gaussian profile of 700 meV width (in the case of Ar atom) and 1000 meV width (in the case of OCS molecule) to simulate the spectrometer resolution [17, 25].

In Fig. 1 we show calculated photoelectron spectra together with the experimental results [17] for photoionization of Ar atom at the excess photon energy above the threshold $\Delta E = 2.0$ eV and $\Delta E = 4.0$ eV, respectively. In addition, we show the photoelectron spectra calculated within the effective width model for comparison.

All spectra demonstrate the notable PCI distortion. Their maxima are red shifted against the nominal energy values of ΔE and line shapes become asymmetrical. Both the measurement and the calculations reveal these features. Earlier this fact was established for the single and multiple Auger decay processes [1 – 3]. Now this PCI distortion manifests itself in the SRAD processes too.

Fig. 1, a shows an excellent agreement between the measured photoelectron spectrum and calculation results of quantum mechanical approach presented here. The effective width model curve demonstrates poorer agreement with experimental spectrum. The effective width model noticeably underestimates the line shape width. Both theoretical curves show correct PCI energy shift $\Delta E_{ph} = -0.27$ eV of line shape maximum against the photon excess energy $\Delta E = 2.0$ eV. According to Fig. 1, b the PCI energy shift $\Delta E_{ph} = -0.22$ eV at the photon excess energy $\Delta E = 4.0$ eV is 50 meV lower than that at $\Delta E = 2.0$ eV. The higher the photon energy, the higher the photoelectron velocity becomes and, consequently, the PCI takes place at longer distances.

The PCI weakens with increasing interparticle distances. Therefore, PCI effects decrease at higher excess photon energies. Fig. 1, *b* shows that the quantum mechanical approach presented here provides better agreement with the experimental spectrum than the effective width model at $\Delta E = 4.0$ eV too. Though this agreement is not so perfect as that at $\Delta E = 2.0$ eV.

In Fig. 2 we compare the theoretical results with experimental spectrum measured for the $1s$ shell ionization of S atom in the OCS molecule [25] at the excess photon energy $\Delta E = 4.4$ eV. The black circles in Fig. 2 present the measured photoelectron spectrum recorded in coincidence with the molecular ion OCS^{2+} and the molecular fragments $\text{CO}^+ + \text{S}^+$. Similarly to the ionization of argon atom the consistent quantum many-body approach provides here better agreement with the experimental spectrum than that resulted from the effective width model. The line shape of the effective width model is narrower and somewhat blue shifted in comparison to the results of quantum many-body approach. The PCI energy shift $\Delta E_{ph} = -0.24$ eV of the photoelectron line shape provided by the quantum many-body approach is greater than that in the effective width model $\Delta E_{ph} = -0.14$ eV and correlates better with the experimental spectrum.

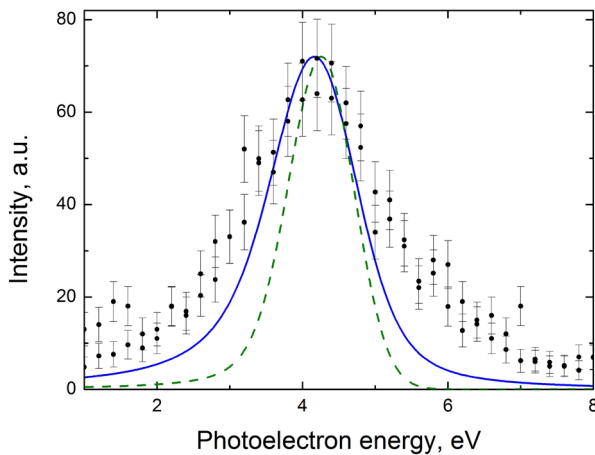


Fig. 2. The photoelectron energy spectrum in $1s^2$ photoinization of S atom in the OCS molecule followed by the SRAD process for the excess photon energy $\Delta E = 4.4$ eV. Black circles present the measured values [25]; the blue solid line shows the photoelectron spectrum calculated within quantum many-body approach according to Eqs.(2) – (7); the green dashed line presents the results of the effective width model

of the effective width model is partly compensated by the low energy resolution of electron spectrometers used in the experiments [17, 25] 0.7 eV and 1.0 eV, respectively. Convolution of the effective width model line shape with the Gaussian profile of Full Width at Half Maximum (FWHM) that equals to the energy resolution makes the results of the effective width model much closer to the experimental spectrum. It is worth to note, that the broadening of the effective width model line shape increases its PCI energy shift. Indeed, in the case of the argon ionization, the PCI energy shifts provided by the effective width model without convolution amount to $\Delta E_{ph} = -150$ meV and $\Delta E_{ph} = -80$ meV for $\Delta E = 2.0$ eV and $\Delta E = 4.0$ eV, respectively.

These values are considerably smaller than the PCI energy shifts of the convoluted effective width curves for the considered excess energies $\Delta E_{ph} = -270$ meV and $\Delta E_{ph} = -220$ meV, respectively, which are quite close to the experimental findings.

According to the Figs. 1, 2 the main shortcoming of the effective width model is that it underestimates systematically the photoelectron line width. It happens because in the SRAD processes in Ar and S atoms the radiative decay width Γ_1 is much larger than the width Γ_2 of the Auger decay. Indeed, in the argon atom the widths of $1s$ and $2p$ vacancies are equal to 0.69 eV and 0.118 eV, respectively. In the S atom their ratio is even higher: $\Gamma_1 = 600$ meV and $\Gamma_2 = 60$ meV. In Ref. [16] it was demonstrated that the line width of the photoelectron spectrum is determined to more extend by the radiative decay width Γ_1 while the Auger electron line width is closer to the Auger decay width Γ_2 . On the other hand, the value of the effective width $\Gamma_{eff} = \Gamma_1 \Gamma_2 / (\Gamma_1 + \Gamma_2)$ in the case of $\Gamma_1 \gg \Gamma_2$ is closer to the smallest one, Γ_2 . For the argon atom the effective width model gives $\Gamma_{eff} = 101$ meV versus $\Gamma_{1s} = 690$ meV and in the OCS case $\Gamma_{eff} = 55$ meV versus $\Gamma_{1s} = 600$ meV. That is why the effective width model fails to reproduce correctly the photoelectron spectrum. This shortcoming



Conclusion

The *ab initio* quantum many-body approach developed in Ref. [16] has been applied for calculations of energy spectra of photoelectrons emitted in 1s shell photoionization of Ar atom and S atom in OCS molecule followed by the SRAD process. The comparison of calculation results with experimental spectra obtained earlier by non-coincidence photoelectron measurements demonstrates quite well agreement. We report a prominent PCI distortion of photoemission energy spectra associated with the SRAD photoionization process. We have also compared the theoretical and experimental spectra with the results of the effective width model. Our calculations demonstrate that the quantum many-body approach provides much more accurate description of the considered photoemission spectra than the approximate model of the effective width. Applying to the photoelectron spectrum the effective width model underestimates considerably its line width and the more consistent quantum many-body approach is needed for the correct description of electron emission resulting from the inner shell photoionization followed by the SRAD process.

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