СЕКЦИЯ 2
РАДИАЦИОННЫЕ ЭФФЕКТЫ В ТВЕРДОМ ТЕЛЕ

SECTION 2
RADIATION EFFECTS IN SOLIDS

REEMISSION OF ENERGY BY IONIZED ATOMS DURING THE CASCADE DECAY OF INNER-SHELL VACANCIES

A.G. Kochur, A.P. Chaynikov, A.I. Dudenko
Rostov State Transport University,
2 Narodnogo Opolcheniya Sq., 344038 Rostov-na-Donu, Russia,
agk@rgups.ru, chaynikov.a.p.@yandex.ru, lesha_11@mail.ru

An atom ionized in its inner electron subshell relaxes to final multiply charged ion states *via* the cascades of consecutive radiative and non-radiative transitions. Only a part of the energy acquired by the atom in the ionization act rests with the atom, a large amount of this energy can be reemitted into the medium by the cascade electrons and photons. The portions of energy (*a*) absorbed by the ionized atom itself, (*b*) reemitted with cascade electrons, and (*c*) reemitted with cascade photons are calculated for the iron, silver and gold atoms singly ionized in their inner to subvalence electron shells. Cascade decay simulations are performed using the method of construction and analysis of the cascade decay trees. The energy redistribution channel *a* is found to be significant only upon ionization of shallow electron subshells in the UV to soft X-ray ranges. The principal channel is *b* making up to 80% of the acquired energy. In Ag and Au, channel *c* is dominant upon *K*-ionization (76% and 89%, respectively). Due to small mean inelastic free paths of the cascade-produced electrons in organism tissues, they play a dominant role in localized energy transfer to the tumor cells when high-Z atoms are used as radiosensitizers in radiotherapy.

Keywords: ionization; cascade relaxation; energy reemission; radiosensitization.

Introduction

Ionization of an inner electron shell of an atom creates a high-energy ionic state which is short-lived; it rapidly decays to lower-in-energy ionic states. Normally, such decays are the cascades of consecutive radiative and non-radiative (Auger, Coster-Kronig, super-Coster-Kronig) transitions [1]. Each cascade transition leads to the emission of either a photon or an electron which carry away the energy acquired by the atom in the ionization act. It is of interest to lean in what proportions the acquired energy is split between the ionized atom itself (channel *a*), and the energies reemitted with cascade electrons (channel *b*) and photons (channel *c*). This information is relevant, for example, in radiotherapy when heavy atoms and nanoparticles made of heavy atoms are used as radiosensitizers, the agents increasing the energy absorption from incident photon or particle beams [2].

In this work we calculate the spectra of the cascade-produced electrons and photons emitted upon the decay of single initial vacancies in the Fe, Ag, and Au atoms, and the energies redistributed through channels *a*, *b* and *c*.

Method of calculation

To simulate a cascade decay of a vacancy, a decay tree must be formed. In the decay tee, the branching points are ionic configurations: the initial-inner-shell-vacancy one, and all the configurations appearing after each consecutive cascade decay step. In contrast to popular Monte-Carlo approach (see, e.g. [1]), we use the method of construction and analysis of the whole decay tree [3,4].

Let $C^{(0)} = nl_j^{-1}$ be the initial ionic configuration with single inner-shell nl_j vacancy. It can decay into a number of the first-decay step configurations $\{C_i^{(1)}\}$. Now if

some of these still have inner-shell vacancies, they decay further forming the second-decay-step set of configurations $\{C_j^{(2)}\}$, *etc*. The decay stops when after some n^{th} decay step all the ionic configurations have vacancies only in the uppermost electron shells and thus can decay no further. Each branching point in a decay tree is characterized by the branching ratios, *i.e.* relative probabilities of all energetically allowed electron transitions from that point to the next-decay-step ones. When the whole decay tree is formed, one can calculate all the characteristics of the cascade decay, such as the spectra of cascade electrons and photons, probabilities of the final ions formation, reemitted energy, *etc*.

When building cascade decay trees, we use the following approximations

– Mean total energies of the cascade ions and partial transition widths are calculated in the Pauli-Fock approximation [5];

– Dependence of branching ratios on ionic configuration is accounted for by introducing partial radiation width per one initial vacancy and one electron, and partial non-radiative widths per one initial vacancy and a pair of electrons [3, 4];

– Multiplet splitting of initial- and final-state configurations of the cascade transitions is included using global characteristics of energy spectra [6].

Results and discussion

The spectra of electrons and photons emitted during the cascade decay of inner-shell vacancies have rather complex multi-component structures since the electron transitions take place in a multitude of different ionic configurations appearing during the cascade development. As an example, Fig. 1 shows the spectra of electrons and photons emitted upon decay of the 1s-vacancy in the gold atom.

The energy acquired by an atom upon nl_j-subshell ionization is equal to the nl_j subshell ionization threshold energy $I(nl_j)$. Mean energies carried away by cascade electrons $E_{\text{out}}^{\text{el}}(nl_j)$ and photons $E_{\text{out}}^{\text{phot}}(nl_j)$ can be calculated using electron and photon spectra of respective nl_j-initial-vacancy cascades:

$$E_{\text{out}}^{\text{el}}(nl_j) = \sum_i E_i P_i^{\text{el}}(nl_j) \quad (1)$$

$$E_{\text{out}}^{\text{el}}(nl_j) = \sum_i E_i P_i^{\text{phot}}(nl_j) \quad (2)$$

Here $P_i^{\text{el}}(nl_j)$ and $P_i^{\text{phot}}(nl_j)$ are the probabilities of non-radiative and radiative transitions during the decay of an nl_j vacancy, E_i are respective electron or photon energies, and the summation is performed over all possible cascade transitions.

The energy absorbed by an ionized atom itself is then

$$E_{\text{abs}}(nl_j) = I(nl_j) - E_{\text{out}}^{\text{el}}(nl_j) - E_{\text{out}}^{\text{phot}}(nl_j) \quad (3)$$

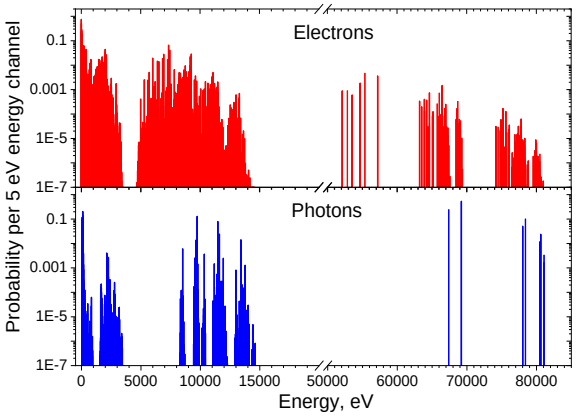

Fig. 1. Spectra of the cascade electrons and photons emitted during the cascade decay of the 1s-vacancy in the gold atom [7]

Fig. 2 shows the relative weights of the energy redistribution channels *a*, *b*, and *c*, *i.e.*

$$x_{\text{abs}}(nl_j) = \frac{E_{\text{abs}}(nl_j)}{I(nl_j)} 100\%,$$

$$x_{\text{out}}^{\text{el}}(nl_j) = \frac{E_{\text{out}}^{\text{el}}(nl_j)}{I(nl_j)} 100\%, \quad (4)$$

$$x_{\text{out}}^{\text{phot}}(nl_j) = \frac{E_{\text{out}}^{\text{phot}}(nl_j)}{I(nl_j)} 100\%,$$

upon the decay of single initial vacancies in the electron subshells of Fe, Ag, and Au.

As can be seen from Fig. 2, the energy absorbed by an ionized atom itself makes significant part of the acquired energy only upon ionization of *M* shell of Fe, *N* shell of

Ag, and *O* and *N* shells in Au. Energy carried away by photons is dominant after *K* ionization of Ag and Au, and noticeable after *K* ionization of Fe and *L* ionization of Au.

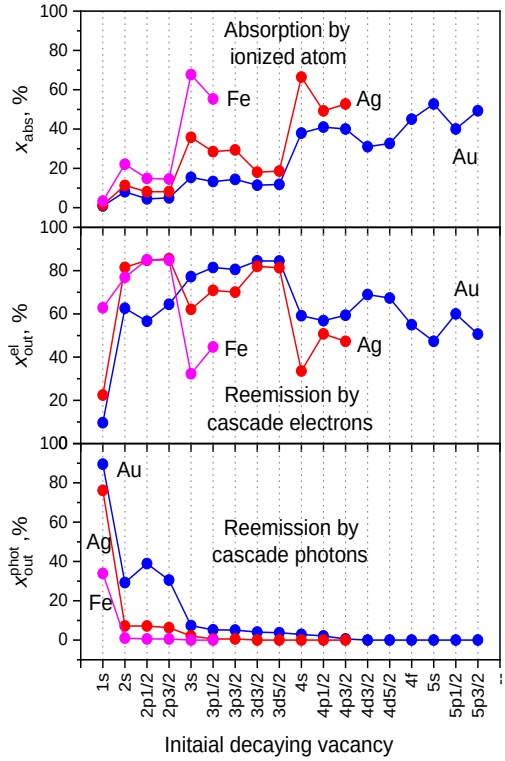

Fig. 2. Relative weights of the acquired energy redistribution channels upon the cascade decay of single vacancies in Au, Ag, and Fe atoms

The principal acquired energy redistribution channel is *b*, *i.e.* reemission of energy by cascade electrons. The cascade electrons are then principal transmitters of energy to the tumor cells when heavy biologically neutral atoms are used as radiosensitizers in radiotherapy of malignant tumors.

Inelastic mean free paths (IMFP) of the electrons produced by the cascade relaxation of vacancies in the gold atom were calculated in [8] as function of incident photon energies. In the incident photon energy range of 0.1 to 1000 keV, IMFP turned out to be 2-7 nm. This is much less than a typical organism cell size meaning that the energy will be transmitted with a high absorbed dose to a particular cell where radiosensitizing atom is placed.

As for the cascade photons, their free paths in organic matter are much larger than the cell size. They will transfer energy to the medium in much larger volumes and with much smaller absorbed dose.

Conclusion

The results of this study are relevant for developing the strategies of radiotherapy using Fe, Ag or Au atoms as radiosensitizing agents.

Acknowledgement

This study was supported by the Russian Science Foundation (project № 23-22-00222, https://rscf.ru/project/23-22-00222/).

References

1. Carlson T.A., Hunt W.E., Krause M.O. Relative abundances of ions formed as the result of inner-shell vacancies in atoms. *Phys. Rev.* 1966; 151(1): 41-47.
2. Liu Y., Zhang P. Li F. Jin X., Li, J., Chen, W., Li Q. Metal-based NanoEnhancers for future radiotherapy: radiosensitizing and synergistic effects on tumor cells. *Theranostics* 2018; 8(7): 1824-1849.
3. Kochur A.G., Dudenko A.I., Sukhorukov V.L., Petrov I.D. Direct Hartree–Fock calculation of multiple Xe^{i+} ion production through inner shell vacancy de-excitations. *J. Phys. B At. Mol. Opt. Phys.* 1994; 27: 1709-1721.
4. Kochur A.G., Sukhorukov V.L., Dudenko A.J., Demekhin P.V. Direct Hartree–Fock calculation of the cascade decay production of multiply charged ions following inner-shell ionization of Ne, Ar, Kr and Xe. *J. Phys. B At. Mol. Opt. Phys.* 1995; 28: 387-402.
5. Kau R., Petrov I.D., Sukhorukov V.L., Hotop H. Experimental and theoretical cross sections for photoionization of metastable Xe* (6s 3P2, 3P0) atoms near threshold. *Zeitschrift für Phys. D Atoms, Mol. Clust.* 1997; 39: 267–281.
6. Kučas S., Karazija R. Global characteristics of atomic spectra and their use for the analysis of spectra. I. energy level spectra. *Phys. Scr.* 1993; 47: 754-764.
7. Chaynikov A.P., Kochur A.G., Dudenko A.I., Petrov I.D., Yavna V.A. Final ion charge spectra upon cascade decay of inner-shell vacancies in atomic Au. *Phys. Scr*. 2023; 98: 025406.
8. Chaynikov A.P., Kochur A.G., Dudenko A.I., Yavna V.A. Cascade energy reemission after inner-shell ionization of atomic gold. Role of photo- and cascade-produced electrons in radiosensitization using gold-containing agents. *J. Quant. Spectrosc. Radiat. Transf.* 2023; 302: 108561.