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Cascade model of X-ray M emission for the atoms of heavy metals

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Cascade model of X-ray M emission for the atoms of heavy metals is proposed that allows one to take into account the main channels of vacancy transfer from L to M electronic subshells, which are responsible for the generation of double-vacancy ($M_{4,5}N$ and $M_{4,5}O$) and triple-vacancy ($M_{4,5}N^2$, $M_{4,5}NO$, and $M_{4,5}O^2$) states. The model allows to separately calculate the contributions of L and M subshells in the emission cross sections of M_5N and M_4N satellites, as well as $M\alpha_{1,2}$ lines along with M_5O satellites and $M\beta$ line with M_4O satellites. Comparison of relative emission cross sections of the main components of $M\alpha$ and $M\beta$ atomic spectra of Au with the experimental relative intensities of these components, excited by $K\alpha_{1,2}$ radiation of Cu and Mo, indicates the correctness of the cascade model used.

Предложена каскадная модель рентгеновской M эмиссии для атомов тяжелых металлов, позволяющая учесть основные каналы переноса вакансий из L в M электронные подоболочки, в результате которых генерируются двухвакансионные ($M_{4,5}N$ и $M_{4,5}O$) и трехвакансионные ($M_{4,5}N^2$, $M_{4,5}NO$ и $M_{4,5}O^2$) состояния. Модель позволяет отдельно рассчитать вклады L и M подоболочек в сечения излучения M_5N и M_4N сателлитов, а также $M\alpha_{1,2}$ линий совместно с M_5O сателлитами и $M\beta$ линии с M_4O сателлитами. Сравнение относительных сечений излучения основных компонентов $M\alpha$ и $M\beta$ спектров атомов Au с экспериментальными относительными интенсивностями этих компонент при возбуждении $K\alpha_{1,2}$ излучением Cu и Mo свидетельствует о корректности использованной каскадной модели.

1. Introduction

Thin films of heavy metals, in particular, Au, are widely used in modern microelectronic devices. Particular important for these films is the control of the films physical state which is mainly defined by the state of electronic subsystem of such structures. Among effective diagnostic methods of the state of electronic subsystem of the metal films, X-ray emission spectroscopy methods of K and L bands are worth to be noted. These methods allow to obtain data of electronic states density in particular, near the Fermi level [1–3]. However, X-ray M spectroscopy was not used for investigation of thin films of heavy metals. At the same time, in order to develop the new materials for protection from a radiation as well as to develop X-ray emitters with certain properties it should properly take into account the X-ray of M series, which in the case of elements with an atomic number $Z > 70$ is already enough intensive.

The X-ray M emission spectrum contains three most intense groups of lines emitted upon radiative filling of the initial M_5, M_4, and M_3 vacancies by electrons from N subshells, i.e., $M\alpha_{1,2}$, $M\beta$, and $M\gamma$ lines, respectively. It is important to note that, for heavy elements with $Z > 70$, the diagram lines $M\alpha_{1,2}$ (the M_5–$N_{6,7}$ transitions), $M\beta$ (M_4–N_6), and $M\gamma$ (M_3–N_5) are accompanied by the groups of rather intense high energy $M_{4,5}N$ and $M_{4,5}O$ satellites related to the listed one-electron transitions in the presence of one or several additional vacancies in N or O subshells [4]. The main mechanism by which these double-vacancy (M_iN and M_iO) and triple-vacancy (M_iN^2, M_iNO, and M_iO^2) states ($i = 3$–5) are generated is related to Coster-Kronig (CK) transitions M_j–M_iN and M_j–M_iO ($i > j$) because their yields for M subshells of atoms with $Z > 70$ are significant and their sums for each of M_1, M_2 and M_3 subshell of, e.g., Au, reach 0.93, 0.90, and 0.83, respectively [5]. In a number of studies, such relative integrated intensities of $M\alpha_{1,2}$, $M\beta$, and $M\gamma$ lines of the heavy elements were measured experimentally under various excitation conditions, for example, at photoabsorption (elements studied are Au, Pb, Th, U [5], and Bi [6]), bombardment by α-particles (Hf–Th) [7, 8], electron (Au, Bi) [9] and proton impacts (Hg, Th, U) [10, 11], and bombardment by ions of F, C, Li (Au) [12]. The obtained values of relative intensities were usually compared with calculated emission cross sections of $M\alpha_{1,2}$, $M\beta$, and $M\gamma$ lines, in which the total yields of the CK transitions and their cascades were taken into account, but the contributions of the CK transitions M_j–M_iN and M_j–M_iO were not separated. In addition, the relative intensities of $M_{4,5}N$ and $M_{4,5}O$ satellites in these papers are not separately determined. In its turn, the use of relative intensities of separated high energy satellites allows one to extend the number of equations describing the intensity of X-ray M emission, thus increasing the informative value of such a description. Similarly, in the case of spectra of L series of metals W, Re, Os, Ir, Pt, the consideration of the relative intensities of separated L_3M satellites of $L\alpha_{1,2}$ and $L\beta_{2,15}$ lines has allowed us to determine the partial width of L_1 level connected with the CK transitions L_1–L_3M_5 [13].

It should be noted that the proposed models of X-ray M emission [14, 15] are restricted to the absorbed photon energies below the ionization threshold of L subshells, which corresponds to the allowance for the generation and migration of vacancies only in M subshells. At the same time, a calculation of the emission cross sections of the high energy satellites of M series at the energies of ionizing particles (photons, electrons, and ions) above the ionization threshold of L subshells would by useful for investigating both X-ray emission and ionization cross sections of M subshells of the heavy elements in a rather wide energy range of ionizing particles.

Therefore in the present paper the cascade model of X-ray M emission which takes into account the basic channels of migration of vacancies from L to M electronic subshells has been proposed that allows calculating the emission cross sections of the main components of the $M\alpha$ and $M\beta$ atomic spectra of the heavy metals with $Z > 70$.

2. The equations describing X-ray M emission

We assume that the energy of a projectile is sufficient for ionization of all M and L subshells and consider the possible processes resulted in formation of the states which are initial for the group of high energy M_5N satellites separated from the $M\alpha$ spectrum. In addition to two-vacancy M_5N_i states, we will take into account three-vacancy $M5N_iN_j$ and $M_5N_iO_j$ states, since their radiative decay also gives rise to the high energy satellites. Let the primary vacancy appears in M_1 subshell. Then the following processes are possible: (a) CK transitions M_1–M_5N_i, whose yield is f_{15N}; (b) cascades of two consecutive CK transitions M_1–M_jN_i, M_j–M_5N_k ($j = 2$–4), the yield is $f_{1jN}f_{j5N}$; (c) cascades of two consecutive CK transitions M_1–M_jN_i, M_j–M_5O_k ($j = 2$–4), the yield is $f_{1jN}N\ f_{j5O}$, and M_1–M_jO_i, M_j–M_5N_k ($j = 2$–4), the yield is $f_{1jO}\ f_{j5N}$; (d) cascades of CK transitions M_1–M_jN_i with the following radiative transitions M_j–M_5 (the yield ω_{j5}), cascades yield is $f_{1jN}\ \omega_{j5}$;

(e) cascades of radiative transitions M_1–M_j with the following CK transitions M_j–M_5N_i, the yield is $\omega_{1j}\ f_{j5N}$; (f) the shake-off (SO) process with additional ionization of the one of N_i subshells (the probability $P^{(N)}_{M1}$) and the following CK transitions M_1–M_5N_i, M_1–M_5O_i (the yield of the last ones f_{15O}), and radiative transition M_1–M_5, cascades yields are $P^{(N)}_{M1}f_{15N}$, $P^{(N)}_{M1}\ f_{15O}$, and $P^{(N)}_{M1}\ \omega_{15}$, respectively; (g) SO process with additional ionization of the one of O_i subshells (the probability $P^{(O)}_{M1}$) and the following CK transitions M_1–M_5N_i, the yield is $P^{(O)}_{M1}\ f_{15N}$. The total yield of the processes, as a result of which a vacancy from M_1 subshell migrates into M_5 subshell with formation of the additional vacancies N, N^2, and NO, is following

$$\begin{aligned} F_{M1} = {} & f_{15N} + f_{12N}(f_{25O} + f_{25N} + \omega_{25}) + \\ & + f_{13N}(f_{35O} + f_{35N} + \omega_{35}) + \\ & + f_{14N}(f_{45O} + f_{45N} + \omega_{45}) + \\ & + f_{12O}f_{25N} + f_{13O}f_{35N} + f_{14O}f_{45N} + \\ & + \omega_{12}f_{25N} + \omega_{13}f_{35N} + \omega_{14}f_{45N} + \\ & + P_{M1}^{(N)}(\omega_{15} + f_{15N} + f_{15O}) + P_{M1}^{(O)}f_{15N}. \end{aligned} \tag{1}$$

Analogous equations can also be written for the yields of the processes of vacancy migration from M_2, M_3, and M_4 subshells to M_5 subshell (F_{Mi}). In the case of the direct ionization of M_5 subshell, M_5N satellites can be formed only due to the SO process with emission of N electron (the probability $P_{M5}^{(N)}$).

Then, in M emission model, it is necessary to take into account the processes of vacancy transfer from L to the M subshells. In particular, under the absorbed photon energies $E = 17.479$ keV (Mo $K\alpha_{1,2}$ lines) the role of these processes can be significant because, in this case, the photoionization cross sections of L subshells (σ_{Li}) are 5–10-fold larger than that of M subshells (σ_{Mi}) (for example, for Au under this photon energy, we have $\sigma_{M1} = 1401$ b, $\sigma_{M5} = 877$ b, $\sigma_{L1} = 5712$ b, and $\sigma_{L3} = 12958$ b [16]). Let the primary vacancy appears in L_1 subshell. Then, the creation of the initial states for M_5N satellites can occur via the following channels: (a) Auger transitions L_1–M_5N, whose yield is a_{L1M5N}; (b) cascades of initial Auger transitions L_1–M_iN, subsequent CK transitions M_i–M_5N, M_i–M_5O, and radiative transitions M_i–M_5 (i = 1–4), the yield is $a_{L1MiN}(f_{i5O} + f_{i5N} + \omega_{Mi5}$; (c) cascades of the initial Auger transitions L_1–M_iM_5, subsequent Auger transitions M_i–NN, M_i–NO, and radiative transitions M_i–N (i = 1–5), the yield is $a_{L1MiM5}(a_{MiNO} + a_{MiN} + \omega_{MiN})$; (d) cascades of the initial Auger transitions L_1–M_iO and subsequent CK transitions M_i–M_5N (i = 1–4), the yield is $a_{L1MiO}f_{i5N}$; (e) cascades of initial radiative transitions L_1–M_i and subsequent CK transitions M_i–M_5N (i = 1–4), the yield is $\omega_{L1Mi}f_{i5N}$; (f) the SO process L_1–L_1N (the probability $P_{L1}^{(N)}$) with subsequent Auger transitions L_1–M_5N, L_1–M_5O and radiative transitions L_1–M_5, the yield is $P_{L1}^{(N)}(\omega_{L1M5} + a_{L1M5O} + a_{L1M5N})$; (g) SO process L_1–L_1M_5 (the probability $P_{L1}^{(M5)}$) with the subsequent Auger transitions L_1–N_iN_j, L_1–N_iO_k and radiative transitions L_1–N, the yield is $P_{L1}^{(M5)}(\omega_{L1N} + a_{L1NO} + a_{L1NN})$; (h) SO process L_1–L_1O (the probability $P_{L1}^{(O)}$) with the subsequent Auger transitions L_1–M_5N, the yield is $P_{L1}^{(O)}a_{L1M5N}$; (i) cascades of the initial CK transitions L_1–L_3M_5, subsequent Auger transitions L_3–NN, L_3–NO, and radiative transitions L_3–N, the yield is $f_{L1L3M5}(a_{L3NO} + a_{L3NN} + \omega_{L3N})$; (j) cascades of the initial CK transitions L_1–L_3N, subsequent Auger transitions L_3–M_5N, L_3–M_5O, and radiative transitions L_3–M_5, the yield is $f_{L1L3N}(a_{L3M5O} + a_{L3M5N} + \omega_{L3M5})$; (k) cascades of the initial CK transitions L_1–L_2N, subsequent Auger transitions L_2–M_5N, L_2–M_5O, and radiative transitions L_2–M_5, the yield is $f_{L1L2N}(a_{L2M5O} + a_{L2M5N} + \omega_{L2M5})$; (l) cascades of the initial CK transitions L_1–L_3O, subsequent Auger transitions L_3–M_5N, the yield is $f_{L1L3O}a_{L3M5N}$; (m) cascades of the initial CK transitions L_1–L_2O and subsequent Auger transitions L_2–M_5N, the yield is $f_{L1L2O}a_{L2M5N}$.

Note that Auger transitions L_1–M_iM_5 and CK transitions L_1–L_3M_5 are accompanied by the formation of double vacancy states M_iM_5, L_3M_5, whose subsequent decay may be related to the filling of M_5 vacancy in the presence of M_i or L_3 vacancies; these processes eliminate the M_iM_5, L_3M_5 states from the subsequent cascades of M_5N satellites formation. They can be taken into account using the rearrangement coefficients $R_{i5} = \Gamma_{M5}/(\Gamma_{Mi} + \Gamma_{M5})$ and $R_{L3M5} = \Gamma_{M5}/(\Gamma_{L3} + \Gamma_{M5})$, i = 1–5. Then, the total yield of the processes responsible for the vacancy migration from L_1 to M_5 subshell with the formation of additional N, N^2, and NO vacancies is equal to

$$\begin{aligned} & F_{L1} = \\ & = a_{L1M5N} + P_{L1}^{(N)}(\omega_{L1M5} + a_{L1M5O} + a_{L1M5N}) + \\ & + P_{L1}^{(M5)}(\omega_{L1N} + a_{L1NO} + a_{L1NN}) + \\ & + P_{L1}^{(O)}a_{L1M5N} + \sum_{i=1}^{4} a_{L1Mi5N}(f_{i5O} + f_{i5N} + \omega_{Mi5}) + \\ & + \sum_{i=1}^{5} a_{L1MiM5}(1 - R_{i5})(a_{MiNO} + a_{MiNN} + \omega_{MiN}) + \\ & + \sum_{i=1}^{4} a_{L1MiO}f_{i5N} + \sum_{i=1}^{4} \omega_{L1Mi}f_{i5N} + \\ & f_{L1L3M5}(1 - R_{L3M5})(a_{L3NO} + a_{L3NN} + \omega_{L3N}) + \end{aligned}$$

$$f_{L1L3N}(a_{L3M5O} + a_{L3M5N} + \omega_{L3M5}) + + f_{L1L3O}a_{L3M5N} + + f_{L1L2N}(a_{L2M5O} + a_{L2M5N} + \omega_{L2M5}) + + f_{L1L2O}a_{L2M5N}. \quad (2)$$

A similar expression can also be written for the yields of the processes of vacancy migration from L_2 and L_3 subshells to M_5 subshell (F_{L2} and F_{L3}). Using the migration coefficients of vacancies in M_i subshells (F_{Mi}), we can present the emission cross section of M_5N satellites with an allowance for the ionization of L_i subshells in the form

$$\sigma_{\alpha S} = \frac{k_\alpha \Gamma_\alpha^R}{\Gamma_{M5}}(\sigma_{M5}P_{M5}^{(N)} + + \sum_{i=1}^{4}\sigma_{Mi}F_{Mi} + \sum_{i=1}^{3}\sigma_{Li}F_{Li})(1 - R_\alpha), \quad (3)$$

where Γ_α^R is the portion of M_5 level width that corresponds to the radiative transition M_5–$N_{6,7}$. We may assume that the radiative transition width Γ_α^R does not change with the appearance of an additional vacancy [17]. The coefficient $k_\alpha = l3/l4$ takes into account the decrease in the total probability of the radiative transition M_5–$N_{6,7}$ in the presence of $N_{6,7}$ vacancy. The rearrangement coefficient R_α determines the relative number of atoms in which $N_{6,7}$ vacancy decays in the presence of M_5 vacancy, $R_\alpha = \Gamma_{N6,7}/(\Gamma_{N6,7} + \Gamma_{M5})$, where Γ_{M5} and $\Gamma_{N6,7}$ are the total widths of M_5 and $N_{6,7}$ levels. This process converts $M_5N_{6,7}$ and $M_5N_{6,7}O$ states into the M_5O, M_5O^2 states, which are initial for M_5O satellites.

Reasoning in the similar way, we can obtain the equation for the yield of the processes of migration of M_1 vacancy into M_5 subshell (G_{M1}), and the equation for the yield of the processes of migration of L_1 vacancy into M_5 subshell (G_{L1}), which are accompanied with the formation of two-vacancy M_5O and three-vacancy M_5O^2 states initial for M_5O satellites

$$G_{M1} = f_{15O} + \omega_{15} + + f_{12O}(f_{25O} + \omega_{25})f_{13O}(f_{35O} + \omega_{35})f_{14O}(f_{45O} + \omega_{45}) + + \omega_{12}f_{25O} + \omega_{13}f_{35O} + \omega_{14}f_{45O} + + P_{M1}^{(O)}(\omega_{15} + f_{15O}), \quad (4)$$

$$G_{L1} = a_{L1M5O} + P_{L1}^{(O)}(a_{L1M5O} + \omega_{L1M5}) + + P_{L1}^{(M5)}\omega_{L1O} + \sum_{i=1}^{4}a_{L1MiO}(f_{i5O} + \omega_{Mi5}) + + \sum_{i=1}^{5}a_{L1MiM5}(1 - R_{i5})\omega_{MiO} + + f_{L1L3M5}(1 - R_{L3M5})\omega_{L3O} + + f_{L1L3O}(\omega_{L3M5} + a_{L3M5O}) + + f_{L1L2O}(\omega_{L2M5} + a_{L2M5O}). \quad (5)$$

The equations for the yields of such processes of vacancy migration from M_2, M_3, M_4 subshells into M_5 subshell (G_{Mi}) and the equations for the yields of such processes of vacancy migration from L_2, L_3 subshells into M_5 subshell (G_{Li}) are similar to that presented above. Then the emission cross section of $M\alpha_{1,2}$ lines together with M_5O satellites is equal to

$$\sigma_\alpha = = \frac{\Gamma_\alpha^R}{\Gamma_{M5}}\left(\sigma_{M5}(1 - P_{M5}^{(N)} + \sum_{i=1}^{4}\sigma_{Mi}G_{Mi} + \sum_{i=1}^{3}\sigma_{Li}G_{Li}\right) + + \sigma_{\alpha S}\frac{R_\alpha}{1 - R_\alpha}, \quad (6)$$

We can also easily take into account the contributions of M and L subshells to the emission cross sections of M_4N satellites and $M\beta$ line together with M_4O satellites. The corresponding emission cross sections are equal to

$$\sigma_{\beta S} = \frac{k_\beta \Gamma_\alpha^R}{\Gamma_{M4}}(\sigma_{M4}P_{M4}^{(N)} + \sum_{i=1}^{3}\sigma_{Mi}H_{Mi} + + \sum_{i=1}^{3}\sigma_{Li}H_{Li})(1 - R_\beta), \quad (7)$$

$$\sigma_\beta = = \frac{\Gamma_\beta^R}{\Gamma_{M4}}\left(\sigma_{M4}(1 - P_{M4}^{(N)}) + \sum_{i=1}^{3}\sigma_{Mi}K_{Mi} + \sum_{i=1}^{3}\sigma_{Li}K_{Li}\right) + + \sigma_{\beta S}\frac{R_\beta}{1 - R_\beta}, \quad (8)$$

where H_{Mi}, K_{Mi}, and H_{Li}, K_{Li} are the yields of the processes of migration of M_i and L_i vacancies into the M_4 subshell, $R_\beta =$

Table 1. Contributions of M and L subshells to the emission cross sections of M_5N satellites of Au (normalized to $\sigma_{\alpha S}$)

$\sigma_{\alpha S}(M_1)$	$\sigma_{\alpha S}(M_2)$	$\sigma_{\alpha S}(M_3)$	$\sigma_{\alpha S}(M_4)$	$\sigma_{\alpha S}(M_5)$	$\sigma_{\alpha S}(L_1)$	$\sigma_{\alpha S}(L_2)$	$\sigma_{\alpha S}(L_3)$	$\sigma_{\alpha S}(M)$	$\sigma_{\alpha S}(L)$
0.10	0.04	0.22	$6{\cdot}10^{-5}$	$2{\cdot}10^{-3}$	0.08	0.15	0.41	0.36	0.64

Table 2. Contributions of M and L subshells to the emission cross sections of $M\alpha_{1,2}$ lines with M_5O satellites of Au (normalized to σ_α)

$\sigma_\alpha(M_1)$	$\sigma_\alpha(M_2)$	$\sigma_\alpha(M_3)$	$\sigma_\alpha(M_4)$	$\sigma_\alpha(M_5)$	$\sigma_\alpha(L_1)$	$\sigma_\alpha(L_2)$	$\sigma\alpha(L_3)$	$\sigma_\alpha(R)$
$6{\cdot}10^{-4}$	$3{\cdot}10^{-3}$	0.05	0.01	0.23	0.01	0.04	0.15	0.51

Table 3. Contributions of M and L subshells to the emission cross sections of M_4N satellites of Au (normalized to $\sigma_{\beta S}$)

$\sigma_{\beta S}(M_1)$	$\sigma_{\beta S}(M_2)$	$\sigma_{\beta S}(M_3)$	$\sigma_{\beta S}(M_4)$	$\sigma_{\beta S}(L_1)$	$\sigma_{\beta S}(L_2)$	$\sigma_{\beta S}(L_3)$	$\sigma_{\beta S}(M)$	$\sigma_{\beta S}(L)$
0.08	0.28	0.06	$3{\cdot}10^{-3}$	0.11	0.27	0.20	0.42	0.58

Table 4. Contributions of M and L subshells to the emission cross sections of $M\beta$ line with M_4O satellites of Au (normalized to σ_β)

$\sigma_\beta(M_1)$	$\sigma_\beta(M_2)$	$\sigma_\beta(M_3)$	$\sigma_\beta(M_4)$	$\sigma_\beta(L_1)$	$\sigma_\beta(L_2)$	$\sigma_\beta(L_3)$	$\sigma_\beta(R)$
$7{\cdot}10^{-4}$	0.02	$4{\cdot}10^{-5}$	0.28	0.02	0.20	0.04	0.44

$\Gamma_{N6,7}/(\Gamma_{N6,7}+\Gamma_{M4}$, Γ_β^R is the width of M_4–N_6 radiative transition, and $k_\beta = 5/6$.

Emission cross sections (3), (6), (7), (8) of Au were calculated for photon energies in the range of $E = 5$–30 keV. We used the photoionization cross sections from [16], the yields of CK transitions f_{ijN} and f_{ijO} from [18], the yields of Auger and CK transitions from L subshells a_{LjMiM5}, a_{LjMiN}, a_{LjMiO}, a_{LjNO}, f_{LiLjM}, f_{LiLjN}, f_{LiLjO} from [19], the fluorescence yields ω_{ij} and the radiative transition widths Γ_β^R, and Γ_α^R from [20], the level widths Γ_{M4}, Γ_{M5} and $\Gamma_{N6,7}$ from [21], and the probabilities of SO processes from [22] ($P_{Li}{}^{(N)} = 0.03$, $P_{Li}{}^{(O)} = 0.16$, $P_{Li}{}^{(M5)} = 10^{-3}$, $P_{Mi}{}^{(N)} = 0.20$, $P_{Mi}{}^{(O)} = 0.14$).

3. Results and discussion

Let us estimate the contributions of individual M and L subshells to the emission cross sections $\sigma_{\alpha S}$, σ_α and $\sigma_{\beta S}$, σ_β of Au in the case of excitation by photons with the energy $E = 17.479$ keV (Mo$K\alpha_{1,2}$ lines). The normalized values of these partial cross sections are determined by individual terms in formulas (3), (6), (7), (8) and are listed in Tables 1–4. Table 1 shows that the main processes of generation of the initial states of M_5N satellites of Au are Auger and radiative transitions from L_3 subshell. Their contribution to the excitation of M_5N satellites is 41 %, and the total contribution of L subshells is 64 %. At the same time, the contribution of L and M subshells to the excitation of the $M\alpha_{1,2}$ lines together with M_5O satellites (Table 2) does not exceed 49 %. In this case, the main process is the rearragement with the cross section $\sigma_\alpha(R) = \sigma_{\alpha S}R_\alpha/(1-\alpha)$, which results in the converting the initial states of M_5N satellites into the initial states of M_5O satellites. The contribution of these processes is 51 %. Table 3 shows that the main processes of generation of the initial states of M_4N satellites of Au are Auger, Coster-Kronig and radiative transitions from the L_2 and M_2 subshells (27 % and 28 %, respectively). At the same time, the contribution of L subshells to the excitation of the $M\beta$ line together with M_4O satellites (Table 4) does not exceed 26 %, and the contribution of M subshells is almost the same (30 %). In this case, the main process is the rearrangement with the cross section $\sigma_\beta(R) = \sigma_{\beta S}R_\beta/(1-R_\beta)$, which results in the converting the initial states of M_4N satellites into the initial states of M_4O satellites ($\sigma_\beta(R) = 44$ %).

Table 5 presents the ratios of the emission cross sections of M_5N satellites to the emission cross sections of $M\alpha_{1,2}$ lines together with M_5O satellites $\chi = \sigma_{\alpha S}/\sigma_\alpha$, as well as the ratios of the emission cross sections of M_4N satellites to the emission cross sections of $M\beta$ line together with M_4O satellites $\gamma = \sigma_{\beta S}/\sigma_\beta$ of Au excited by $K\alpha_{1,2}$ ra-

Table 5. Ratios of the emission cross sections of components of $M\alpha$ and $M\beta$ spectra of Au

Energy of absorbed photons, keV	χ		γ		η	
	calculation	experiment	calculation	experiment	calculation	experiment
8.048 Cu $K\alpha_{1,2}$	0.72	0.69±0.07	0.58	0.53±0.05	0.68	0.67±0.05
17.479 Mo $K\alpha_{1,2}$	1.56	1.48±0.15	1.28	1.24±0.12	0.59	0.57±0.04

diation of Cu and Mo anodes. In addition, Table 5 presents the ratios of the emission cross sections of the total profile of $M\beta$ and $M\alpha$ lines (including M_4N and M_5N satellites, respectively) $\eta = \sigma_{\beta\sum}/\sigma_{\alpha\sigma}$. The calculated values χ, γ, and η were compared with experimentally determined relative intensities of the corresponding components of Au M spectra (Table 5). The $M\alpha$ and $M\beta$ fluorescence spectra of Au were recorded with a DRS-2 X-ray spectrograph in the first order of reflection from the $(10\bar{1}1)$ planes of a Johann-bent quartz single crystal. Due to a strong self-absorption, the $M\gamma$ spectrum was not recorded. The chemical purity of samples was higher than 99.9 %. The spectra were excited by the monochromatized $K\alpha_{1,2}$ radiation of BSV-29 X-ray tubes with Cu and Mo anodes. The $K\alpha_{1,2}$ lines were separated from the primary polychromatic spectra by a focusing graphite monochromator in the first order of reflection from the (002) planes. The tubes operated with the voltage $U = 30$ kV and the current $I = 30$ mA. It is important that the photons energy of the Mo $K\alpha_{1,2}$ radiation is sufficiently high to ionize L subshells of Au atoms (the photons energies of Cu and Mo $K\alpha_{1,2}$ lines are 8.048 keV and 17.479 keV, respectively, and the ionization potentials of L subshells of Au are $E_{L1} = 14.353$ keV, $E_{L2} = 13.734$ keV and $E_{L3} = 11.919$ keV [23]). More detailed experimental technique of obtaining and processing $M\alpha$ and $M\beta$ fluorescence spectra of Au is presented in our previous paper [24].

Table 5 shows that the calculation reflects experimentally revealed increasing the relative intensities of $M_{4,5}N$ satellites of Au (by a factor of 2.1 for χ and 2.3 for γ) during the transition from $K\alpha_{1,2}$ excitation of Cu (L subshells are not ionized) to $K\alpha_{1,2}$ excitation of Mo. This increase of the values of χ and γ is being caused by appearance of the powerful channels of vacancy migration from L subshells via excitation of Au $M\alpha$ and $M\beta$ spectra by $K\alpha_{1,2}$ radiation of Mo. It is important that the calculated and experimentally determined values of χ, γ, and η of Au coincide well with each other, which proves the correctness of the proposed cascade model of X-ray M emission.

4. Conclusions

Thus, the cascade model of X-ray M emission for the heavy metals atoms is proposed that allows one to take into account the main channels of vacancy transfer from L to M subshells, which are responsible for the generation of double-vacancy ($M_{4,5}N$ and $M_{4,5}O$) and triple-vacancy ($M_{4,5}N^2$, $M_{4,5}NO$ and $M_{4,5}O^2$) states. Comparison of the relative emission cross sections of M_5N and M_4N satellites of Au with the experimental relative intensities of these satellites, excited by $K\alpha_{1,2}$ radiation of Cu and Mo anodes, indicates the correctness of the represented cascade model. The proposed cascade model of X-ray M emission can be also used for determination of the characteristic parameters of electronic subsystem of thin films of heavy metals with $Z > 70$, in particular ionization cross sections of electronic shells and probabilities of autoionization processes.

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Каскадна модель рентгенівської *M* емісії для атомів важких металів

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Запропоновано каскадну модель рентгенівської M емісії для атомів важких металів, що дозволяє врахувати основні канали переносу вакансій з L в M електронні підоболонки, в результаті яких генеруються двовакансійні ($M_{4,5}N$ і $M_{4,5}O$) та трихвакансійні ($M_{4,5}N^2$, $M_{4,5}NO$ і $M_{4,5}O^2$) стани. Модель дозволяє окремо розрахувати внески L і M підоболонок до перерізів випромінювання M_5N і M_4N сателітів, а також $M\alpha_{1,2}$ лінії разом з M_5O сателітами та $M\beta$ лінії з M_4O сателітами. Порівняння відносних перерізів випромінювання основних компонентів $M\alpha$ і $M\beta$ спектрів атомів Au з експериментальними відносними інтенсивностями цих компонентів при збудженні $K\alpha_{1,2}$ випромінюванням Cu і Mo свідчить про корректність використаної каскадної моделі.