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The $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ X-Ray Emission of Aluminium under Electron Impact

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The relative intensities, $\gamma = I(K_\alpha L^1)/I(K_{\alpha_{1,2}})$, $\eta = I(K_\alpha L^2)/I(K_\alpha L^1)$ and $\chi = I(K_\alpha L^3)/I(K_\alpha L^1)$, of x-ray emission $K_{\alpha_{1,2}}$, $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ lines' groups of aluminium are experimentally studied under the excitation by electron impact in the range of accelerating voltages $U = 4.5$–100 kV. The model of K_α x-ray emission has been proposed and takes into account the main channels of multiply ionized $KL^n_{2,3}$ states' decay. As found, the probabilities of creation of $KL_{2,3}$ configuration (P_1), $KL^2_{2,3}$ configuration (P_2) and $KL^3_{2,3}$ configuration (P_3) monotonically decrease when bombarding-electron energy $E > 20$ keV. The observed decrease of the P_1, P_2 and P_3 values can be explained by decreasing of the average energy transferred to the atom in electron–atom collision. The P_2 and P_3 probabilities significantly exceed (by 1.6 to 2.5 times) their values predicted within the shake-off approximation of simultaneous independent ejecting of two and three $2p$ electrons that indicates a significant role of the $2p$–$2p$ electron correlations in $KL^n_{2,3}$ ionization processes. As shown, the $P_{21} = P_2/P_1$ and $P_{31} = P_3/P_1$ ratios are practically constant in the whole range of accelerating voltages and are the parameters characterizing the values of $2p$–$2p$ electron correlations.

Key words: K_α x-ray emission, multiply ionized $KL^n_{2,3}$ states, $2p$–$2p$ electron correlations.

Відносні інтенсивності $\gamma = I(K_\alpha L^1)/I(K_{\alpha_{1,2}})$, $\eta = I(K_\alpha L^2)/I(K_\alpha L^1)$ та $\chi = I(K_\alpha L^3)/I(K_\alpha L^1)$ груп рентґенівських емісійних ліній $K_{\alpha_{1,2}}$, $K_\alpha L^1$, $K_\alpha L^2$ та

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$K_\alpha L^3$ алюмінію експериментально досліджено при електронному бомбуванні у діяпазоні прискорювальних напруг $U = 4{,}5–100$ кВ. Запропоновано модель рентґенівської K_α-емісії, яка враховує основні канали розпаду кратно йонізованих $KL^n_{2,3}$-станів. Встановлено, що ймовірності створення станів $KL_{2,3}$-конфіґурації (P_1), $KL^2_{2,3}$-конфіґурації (P_2) та $KL^3_{2,3}$-конфіґурації (P_3) монотонно зменшуються в області енергій бомбівних електронів $E > 20$ кеВ. Зменшення величин P_1, P_2 та P_3, яке спостерігається експериментально, може бути зумовленим зменшенням середньої енергії, що передається атому при електрон-атомному зіткненні. Ймовірності P_2 та P_3 значно перевищують (від 1,6 до 2,5 разів) відповідні значення, визначені у наближенні миттєвого незалежного вильоту двох і трьох $2p$-електронів, що свідчить про істотну роль $2p–2p$-електронних кореляцій у процесах $KL^n_{2,3}$-йонізації. Показано, що відношення $P_{21} = P_2/P_1$ та $P_{31} = P_3/P_1$ зберігаються практично сталими в усьому діяпазоні прискорювальних напруг і є параметрами, що характеризують величину $2p–2p$-електронних кореляцій.

Ключові слова: K_α рентґенівська емісія, кратно йонізовані $KL^n_{2,3}$-стани, $2p–2p$-електронні кореляції.

Относительные интенсивности $\gamma = I(K_\alpha L^1)/I(K_{\alpha_{1,2}})$, $\eta = I(K_\alpha L^2)/I(K_\alpha L^1)$ и $\chi = I(K_\alpha L^3)/I(K_\alpha L^1)$ групп рентгеновских эмиссионных линий $K_{\alpha_{1,2}}$, $K_\alpha L^1$, $K_\alpha L^2$ и $K_\alpha L^3$ алюминия экспериментально исследованы при электронной бомбардировке в диапазоне ускоряющих напряжений $U = 4{,}5–100$ кВ. Предложена модель рентгеновской K_α-эмиссии, которая учитывает основные каналы распада кратно ионизированных $KL^n_{2,3}$-состояний. Установлено, что вероятности создания состояний $KL_{2,3}$-конфигурации (P_1), $KL^2_{2,3}$-конфигурации (P_2) и $KL^3_{2,3}$-конфигурации (P_3) монотонно уменьшаются в области энергий бомбардирующих электронов $E > 20$ кэВ. Уменьшение величин P_1, P_2 и P_3, наблюдаемое экспериментально, может быть обусловлено уменьшением средней энергии, которая передаётся атому при электрон-атомном столкновении. Вероятности P_2 и P_3 значительно превышают (от 1,6 до 2,5 раза) соответствующие значения, определённые в приближении мгновенного независимого вылета двух и трёх $2p$-электронов, что свидетельствует о существенной роли $2p–2p$-электронных корреляций в процессах $KL^n_{2,3}$-ионизации. Показано, что отношения $P_{21} = P_2/P_1$ и $P_{31} = P_3/P_1$ сохраняются практически постоянными во всем диапазоне ускоряющих напряжений и являются параметрами, которые характеризуют величину $2p–2p$-электронных корреляций.

Ключевые слова: K_α-рентгеновская эмиссия, кратно ионизированные $KL^n_{2,3}$-состояния, $2p–2p$-электронные корреляции.

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1. INTRODUCTION

The ionization of the K shell of an atom by photon or electron may be accompanied by autoionization with the formation of one or more (n)

additional vacancies in the $L_{2,3}$ subshells [1–3]. The same multiply ionized end states also arise upon ion bombardment; however, the direct $KL^n_{2,3}$ ionization mechanism is crucial in that case [4]. The nature of the autoionization process, the main one under photoionization or electrons' impact, is associated with the 'shaking off' of the $2p$ electrons as a result of the fast self-consistent field change upon the occurrence of a K-vacancy ('shake-off'—SO). It is important that the probability of another additional L-vacancy formation by the SO mechanism can be calculated with a good accuracy within the sudden approximation [5–7]. However, even in the simplest case, it was experimentally found that some features of the double $KL_{2,3}$ ionization cannot be explained solely by the SO process. For instance, the cross-section ratio $\sigma(^3P)/\sigma(^1P)$ of the ^3P and ^1P terms for Si and Cr atoms reveals a different near-threshold behaviour during photoabsorption and electron impact that cannot be explained without accounting for the multielectron effects in the atomic core-slow ejected $1s$, $2p$ electrons' system [8, 9]. Presumably, the role of multielectron effects at the formation of one, two, and three additional $L_{2,3}$ vacancies (with configurations of $KL_{2,3}$, $KL^2_{2,3}$, and $KL^3_{2,3}$, respectively) will increase with the multiplicity of the additional $L_{2,3}$ ionization as the electron–electron correlations are the strongest for autoionization electrons from one subshell [10]. Thus, a comparative experimental study of the probabilities of additional $L_{2,3}$ ionization (P_1), $L^2_{2,3}$ ionization (P_2) and $L^3_{2,3}$ ionization (P_3) at different energies of bombarding electrons allow quantifying the magnitude of the $2p$–$2p$ correlation interaction after ionization on the energy of the incident electron (perturbation rate) as well as on the multiplicity of the $KL^n_{2,3}$ ionization. To the best of our knowledge, theoretical calculations of the $KL^n_{2,3}$ ionization probabilities taking into account the post-collision interaction between the slow two and three $2p$ electrons as well as with the atomic core are not available in the literature. Therefore, such an experiment is of great interest.

Experimental information about the P_1, P_2, and P_3 probabilities can be obtained from the relative intensities of the x-ray emission $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ satellite groups, for which the initial $KL_{2,3}$, $KL^2_{2,3}$, and $KL^3_{2,3}$ configurations are as follow: the $K_\alpha L^1$ group corresponds to the $KL_{2,3}$–$L^2_{2,3}$ radiative transitions, the $K_\alpha L^2$ group—the $KL^2_{2,3}$–$L^2_{2,3}$ radiative transitions, and the $K_\alpha L^3$ group—the $KL^3_{2,3}$–$L^4_{2,3}$ radiative transitions [11, 12]. In the present paper, the relative intensities of x-ray emission AlK_α satellite groups $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ are experimentally studied under electron impact in a wide range of incident electron energies.

2. EXPERIMENTAL METHOD

Aluminium was used as a sample for several reasons. It is a metal, in which the interaction of an outgoing $2p$ electron with an atomic core is effectively shielded by conduction electrons. The AlK_α spectrum can be well resolved by quartz crystal-analysers into individual satellite groups corresponding to various ionization multiplicities as well as into individual components within the group. Additionally, relatively weak group lines $K_\alpha L^2$ and $K_\alpha L^3$ of Al are still reasonably detectable using step-by-step scanning with longer accumulation times.

The x-ray emission AlK_α lines were excited by an electron beam in the range of incident electron energies $E = 4.5$–100 kV. The K_α spectrum were registered in the first order of reflection from the $(10\bar{1}1)$ planes of the quartz monocrystal bent according to Johann in the step-by-step regime (with the step sizes of 0.1–0.4 eV); the accumulation time in a point is of 1–50 s. The method of spectrograms processing is given in Refs. [8, 13, 14]. The corrections were done taking into account the angular dependence of the reflection coefficient of the quartz-analyser as well as the spectrometer dispersion upon the transfer from the angle scale to the energy scale. A typical AlK_α spectrum is shown in Fig. 1 ($E = 25$ keV). The relative intensities $\gamma = I(K_\alpha L^1)/I(K_{\alpha_{1,2}})$, $\eta = I(K_\alpha L^2)/I(K_\alpha L^1)$ along with $\chi = I(K_\alpha L^3)/I(K_\alpha L^1)$ were measured as the ratio of the areas under the experimental contours of the $K_{\alpha_{1,2}}$ lines and $K_\alpha L^1$, $K_\alpha L^2$, $K_\alpha L^3$ satellites, which were singled out from the slope of $K_{\alpha_{1,2}}$. The relative error in determining the η and χ values did not exceed 5–8%.

3. RESULTS AND DISCUSSION

It is important to note that not only the radiative transitions associated with the decay of $KL^2_{2,3}$ configurations contribute to the $K_\alpha L^2$ satellite intensity. The $KL_1L_{2.3}$ and KL^2_1 configurations need to be considered as well. Furthermore, for some of the atoms, the states of the $KL^2_{2,3}$ configuration are created bv the $KL_1L_{2,3}-KL^2_{2,3}M$ and $KL^2_1 - KL^2_{2,3}M^2$ Coster–Kronig transitions. It applies to the $KL^3_{2,3}$ configuration as well. Therefore, the contributions of these processes to the intensity of the $K_\alpha L^2$ and $K_\alpha L^3$ Al satellites were evaluated using values of the total widths of the K and L levels [15], the Auger and Coster–Kronig yields [16], and the $2p$ electron SO probability in occurrence of K vacancy [6, 7]. It has established that the maximum contribution of such processes to the corresponding intensity does not exceed 10–12% in case of Al. Therefore, the experimentally determined intensities $I(K_\alpha L^2)$ and $I(K_\alpha L^3)$ were reduced by 11%.Thus, the creation of satellites due to the Auger and Coster–Kronig processes are not considered.

As given above, the relative intensities $I(K_{\alpha_{1,2}})$, $I(K_\alpha L^1)$, $I(K_\alpha L^2)$, and $I(K_\alpha L^3)$ are determined by the equations (to within the constant common factor):

$$I(K_{\alpha_{1,2}}) = \sigma_K (1 - P_1) \frac{\Gamma_\alpha^R}{\Gamma_K} \; (P_1 >> P_2, P_3), \tag{1}$$

$$I(K_\alpha L^1) = \sigma_K P_1 \frac{(5/6)\,\Gamma_\alpha^R}{a_1 \Gamma_K^A + k_1 \Gamma_K^R + \Gamma_L}, \tag{2}$$

$$I(K_\alpha L^2) = \sigma_K P_2 \frac{(2/3)\,\Gamma_\alpha^R}{a_2 \Gamma_K^A + k_2 \Gamma_K^R + 2\Gamma_L}, \tag{3}$$

$$I(K_\alpha L^3) = \sigma_K P_3 \frac{(1/2)\,\Gamma_\alpha^R}{a_3 \Gamma_K^A + k_3 \Gamma_K^R + 3\Gamma_L}. \tag{4}$$

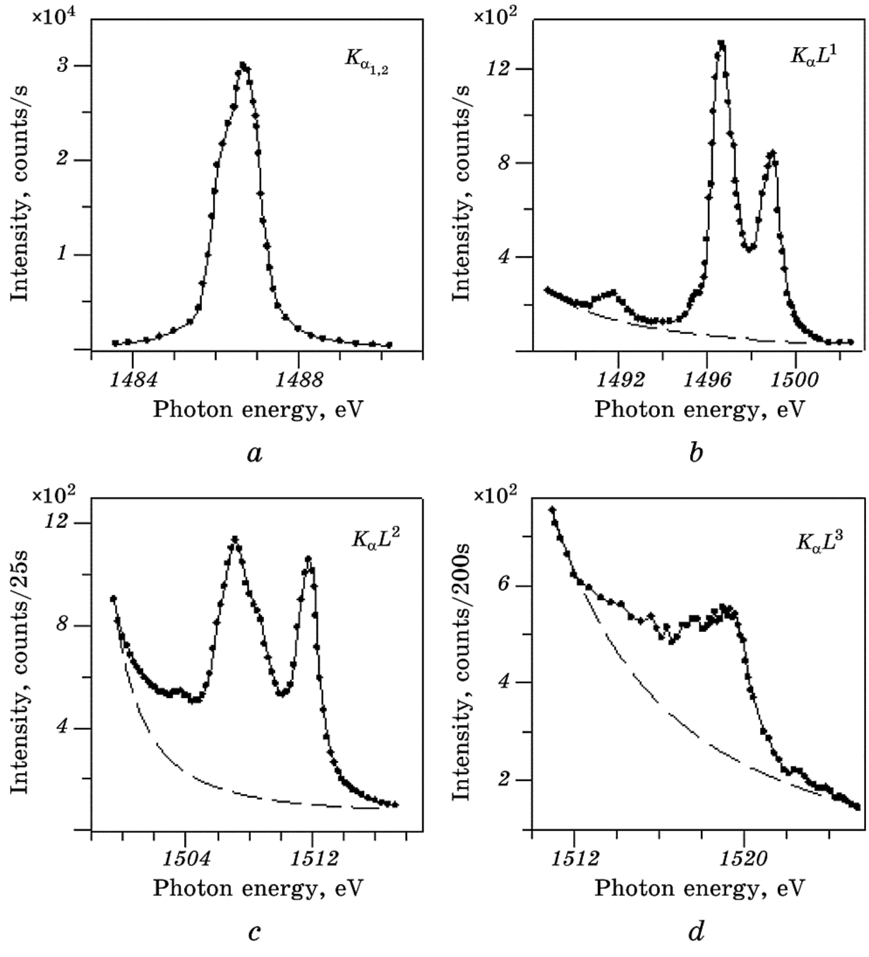

Fig. 1. X-ray K_α spectrum of Al: a—$K_{\alpha_{1,2}}$; b—$K_\alpha L^1$; c— $K_\alpha L^2$; d—$K_\alpha L^3$.

Therefore,

$$P_1 = \frac{\gamma}{\gamma + G},\ P_2 = \eta P_1 N,\ P_3 = \chi P_1 M, \quad (5)$$

$$G = \frac{5}{6}\frac{\Gamma_K}{a_1\Gamma_K^A + k_1\Gamma_K^R + \Gamma_L},\ N = \frac{5}{4}\frac{a_2\Gamma_K^A + k_2\Gamma_K^R + 2\Gamma_L}{a_1\Gamma_K^A + k_1\Gamma_K^R + \Gamma_L},$$

$$M = \frac{5}{3}\frac{a_3\Gamma_K^A + k_3\Gamma_K^R + 3\Gamma_L}{a_1\Gamma_K^A + k_1\Gamma_K^R + \Gamma_L}.$$

In formulas (1)–(5), σ_K—the K shell ionization cross-section, Γ_K and Γ_L—the width of K and $L_{2,3}$ levels, Γ_α^R—the width of the K–$L_{2,3}$ radiative transition, Γ_K^A, Γ_K^R—the Auger and radiative components of the K level width. The coefficients 5/6, 2/3 and 1/2 take into account the reducing of the number of $2p$ electrons with the increasing multiplicity of ionization; a_1, a_2, a_3—the decreasing of Γ_K^A, $k_1 = 5/6$; $k_2 = 2/3$, $k_3 = 1/2$—the Γ_K^R decreasing at the formation of the one, two and three additional $L_{2,3}$ vacancies, respectively. Taking into account the rates of KL_iL_j and KL_iM_j Auger transitions [17], the following parameters were obtained: $a_1 = 0.82$, $a_2 = 0.64$, $a_3 = 0.35$. We used the values as follow: $\Gamma_K = 0.420$ eV, $\Gamma_L = 0.004$ eV [15], the fluorescence yield $\omega_K = 0.039$ [16], resulting in $G = 1.12$, $N = 0.47$, $M = 0.32$.

Thus, the values of the P_1, P_2, and P_3 probabilities were experimentally determined at various incident electron energies based on the relative intensities γ, η, and χ. They are listed in Table 1. The experimental probabilities P_1, P_2, and P_3 demonstrated monotonically decrease with increasing incident electron energy above 20 keV. The decreasing of P_1, P_2, and P_3 with the incident electron energy may be re-

TABLE 1. Parameters of the $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ lines and of the $KL_{2,3}^n$ ionization processes.

Acceleration voltage, kV	Relative intensities			P_1, 10^{-2}	P_2, 10^{-3}	P_3, 10^{-5}	$P_{2,1}$, 10^{-2}	$P_{3,1}$, 10^{-4}
	γ, 10^{-2}	η, 10^{-2}	χ, 10^{-2}					
4.5	10.4	8.3	2.8	8.5	3.3	7.6	3.9	8.9
7	9.9	8.5	3.0	8.1	3.1	7.8	4.0	9.6
11	9.8	8.1	3.1	8.0	3.1	7.7	3.8	9.5
17	9.5	8.2	3.1	7.8	2.9	7.7	3.9	9.9
25	8.8	7.9	3.0	7.3	2.7	6.9	3.7	9.5
50	8.1	8.2	2.9	6.7	2.5	6.1	3.9	9.1
75	7.0	8.3	3.1	5.9	2.2	5.6	3.9	9.5
100	6.5	8.1	3.0	5.5	2.1	5.4	3.8	9.8

lated to the decreased average energy transferred to the atom in the electron–atom collision. Thus, the reduction of the transferred energy should be accompanied by decreasing of the velocity of the ejected 1*s* electron and, in turn, the lowering of the SO processes probabilities.

The experimental values for P_2 and P_3 now will be compared with those calculated in the SO approximation ($P_2^{(SO)}$ and $P_3^{(SO)}$) using the $P_1^{(SO)}$ values [6, 7]. The probability of a simultaneous independent ejection of two arbitrary 2*p* electrons in the occurrence of the *K* vacancy is equal to $P_2^{(SO)} = (5/12)(P_1^{(SO)})^2$ (the probability of ejecting one certain 2*p* electron being $P_1^{(SO)}/6$), for the pair of certain 2*p* electrons, $(1/36)(P_1^{(SO)})^2$, the number of possible pairs is $6!/(2!4!) = 15$. Similarly, in the case of independent simultaneous ejection of three arbitrary 2*p* electrons, the probability is $P_3^{(SO)} = (5/54)(P_1^{(SO)})^3$. For Al atoms, $P_1^{(SO)} = 6.9 \cdot 10^{-2}$ [6, 7], then the corresponding probabilities are $P_2^{(SO)} = 2.0 \cdot 10^{-3}$ and $P_3^{(SO)} = 3.1 \cdot 10^{-5}$. The comparison of the experimental data and the SO estimation shows that in the range of the most effective SO process ($E = 5$–15 keV) the experimental values of the probabilities P_2 and P_3 significantly exceed ones predicted within the approximation of independent 'shaking off', the discrepancy being proportional to the multiplicity of the additional $L_{2,3}$ ionization ($P_2/P_2^{(SO)} = 1.6$; $P_3/P_3^{(SO)} = 2.5$). Therefore, the $KL_{2,3}^2$ and $KL_{2,3}^3$ ionization processes are essentially multielectron, and 2*p*–2*p* electron correlations should be included in the theoretical model of the multiple ionization.

Other parameters of the $KL_{2,3}^n$ ionization processes are the ratios $R_{2,1} = P_2/P_1$ and $R_{3,1} = P_3/P_1$, which determine the relative fractions of atoms with two and three additional $L_{2,3}$ vacancies. Importantly, the values of $R_{2,1}$ and $R_{3,1}$ are constant within the experimental error in the whole range of incident-electrons' energies. It means that $R_{2,1}$ and $R_{3,1}$ do not depend on the perturbation rate and characterize the values of 2*p*–2*p* electron correlations.

4. CONCLUSIONS

The relative intensities γ, η, and χ of Al x-ray emission $K_\alpha L^1$, $K_\alpha L^2$, and $K_\alpha L^3$ satellites under electron bombardment have been experimentally investigated in the energy range $E = 4.5$–100 keV. The x-ray emission model has been proposed that allows taking into account the main channels of creation and decay the states of $KL_{2,3}$, $KL_{2,3}^2$, and $KL_{2,3}^3$ configurations under electron impact. The formation probabilities of one, two and three additional $L_{2,3}$ vacancies P_1, P_2, and P_3 as result of *K* ionization are determined. As established, the values of P_1, P_2, and P_3 monotonically decrease in the range $E > 20$ keV that can be explained by decreasing of the average energy transferred to the atom in the electron–atom collision. The probabilities P_2 and P_3 are significantly exceed (by 1.6 to 2.5 times) the predicted in approximation of simultaneous independent ejecting of two and three 2*p* electrons that indicates a significant role of 2*p*–2*p* electron correlations in $KL^{n}_{2,3}$ ionization processes.

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