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L-subshell fluorescence yields for elements with $73 \leq Z \leq 83$

J. Q. Xu
Shanghai Institute of Nuclear Research, Academia Sinica, P.O. Box 800-204, Shanghai 201849, China
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An analysis of the L x-ray spectra, induced by ~ 50 keV electron impact and recorded with a curved crystal spectrometer, has been performed based on a calculation model that takes into account all physical events taking place during the initial ionization and inner-vacancy processes. The L-subshell fluorescence yields ω_1, ω_2, and ω_3 for ten elements with atomic number $Z = 73-83$ were obtained. They are all in good agreement with recent measurements but display an appreciable disagreement with the semiempirically compiled data for the elements around $Z = 80$. These experimental values of ω_1 are approximately 20% larger than the compiled data and 55% larger than the latest theoretical calculation for the elements with $Z \approx 80$.

I. INTRODUCTION

When the semiempirical compilation of the decay yields of an atomic L-subshell vacancy was given in 1979 by Krause[1] and Krause and Oliver,[2] only a few experimental L_1-subshell fluorescence, Coster-Kronig (CK), and Auger yields and level widths were available. Especially, directly measured yields of the L_1-subshell CK transitions, which are a dominant component of the L_1-vacancy decay if the L_1-$L_{2,3}$ CK channels are open, were nonexistent or inaccurate in most regions of the elemental periodic table. Thus the L_1-subshell values were evaluated mainly based on some theoretical calculations pertaining to singly ionized atoms with some adjustments made to fit experimental results. The L_1-L_2 and L_1-L_3 CK yields, f_{12} and f_{13}, were determined by partitioning the L_1-subshell CK yield f_1 in terms of published theoretical results.

Now the situation is changed. Since that time new data on the L_1-subshell decay yields, directly measured or semiempirically acquired in various primary excitation modes including photo-, electron, and proton ionizations as well as nuclide disintegration, have been reported.[3–16] Even some L_1-$L_{2,3}M$ CK yields, f_{12M} and f_{13M}, for intermediate elements have also been obtained from analyses of the observed satellite and diagram x-ray spectra.[14–16] It has been well known that there is a serious discrepancy between the compiled and theoretical data and the measurements, and experimental information on the L_1-vacancy decay rates is very valuable. In the element region $20 < Z < 30$, the L_1-level widths measured are much larger than those compiled; at $Z = 27$, the measured values are even approximately 4 eV larger than the compiled one.[3] In the intermediate element region, where the L_1-L_2M and L_1-L_3M CK transition cutoffs are located near $Z = 40$ and 50, respectively, the theoretical calculations overestimate the L_1-L_3M CK rates by a factor of about 2.5 (Refs. 4 and 14–17) for the elements whose L_1-L_3M CK processes are energetically allowed, and the cutoff of the L_1-L_2M and L_1-L_3M CK transitions is not sharp with increasing atomic number.[5,14,16,18] The experimental results show that the L_1-L_2M_4 CK transition is forbidden but the L_1-L_2M_5 transition is allowed at $Z = 40$. The complete cutoff for the L_1-L_2M_5 transition is localized at $Z = 41$, for the L_1-L_3M_4 transition at $Z = 48$, for the L_1-L_3M_5 transition at $Z = 52$ in metallic state.[14–16,18] (Please note that the Cd L_1-subshell level width should be $\Gamma_1 = 3.05$ eV in Tables 1 of Refs. 15 and 18, where the two were mistyped.) In the heavy element region, recent measurements of Werner and Jitschin[10] and Marques *et al.*[12] display an obvious disagreement with the theoretical predictions and also with the compilation. On the other hand, we have noticed that experimental data of the L_1-vacancy decay yields for the elements with $Z < 70$ are generally in line with one another,[3,18] whereas those for heavy elements are considerably scattered and still in doubt.

In recent years, we have acquired and reported a large quantity of the L_1-subshell yields of the intermediate elements[14–16] by virtue of the analyses of the satellite and diagram L x-ray spectra recorded with bent-, double-crystal, and photoelectron spectrometers. The success inspires us to study the L-vacancy decay yields of heavy elements in a similar way. But here we directly use L-subshell fluorescence yields rather than both the radiative widths of the L x-ray lines and the level widths of the three L subshells in the calculation model.[14–16] The present L x-ray spectra, induced by electron impact on the elements with $73 \leq Z \leq 83$, were measured by Goldberg[19] with a curved crystal spectrometer, and the relative intensities of the L x-ray satellite and diagram lines were estimated by Ferreira *et al.*[20] and Salgueiro *et al.*[21] Ferreira *et al.* and Salgueiro *et al.* studied the spectra in 1965 and in 1974; these studies led to a measurement of the L_1-subshell CK and fluorescence yields for some of the elements, which was quoted by Bambynek *et al.*,[22] Krause,[1] and others, but we will argue from current knowledge in the next section that their approach and results should be questioned.

In this work, the L x-ray spectra of Goldberg are reanalyzed based on a calculation model of L-vacancy deexcitation processes, which takes into account all physical events taking place during the initial ionization and inner-vacancy decay, and into account the role of possible modifications of the L-subshell fluorescence and CK yields in the case of multiple-vacancy states. This analysis allows us to evaluate the L_2- and L_3-subshell fluorescence yields, ω_2 and ω_3, and to acquire values of the L_1-subshell fluorescence yields, ω_1, for the investigated elements from the experimental L x-ray intensities and the L-subshell ionization cross sections by electron impact.[23] The results of the present study obviously improve the agreement of experimental values of ω_1 for the elements under consideration and display clear disagreement with both the theoretical calculations and the compiled data for the elements around $Z=80$. This work also gives some new estimates of ω_2 and ω_3. They are all in line with recent measurements.

II. BASIS OF THE NEW ANALYSIS

A. Decay yields of heavy elements

Most old measurements and calculations of L-subshell fluorescence and CK yields of heavy elements around $Z=80$, including the work by Ferreira *et al.*,[20] were based on the assumption that L_2-L_3 CK yields $f_{23}=0$.[24–27] In 1955, Ross *et al.*[28] made a comparison between two independent sets of experimental data, one obtained from RaD disintegration and the other from x-ray excitation, and indicated that f_{23} should have a nonzero value for Bi. Sequentially, this proposal was verified by many experiments using high-resolution electron spectrometers[29,30] and Si(Li)/Ge(Li) photon spectrometers with a coincident unit.[31–34] However, those earlier values ($f_{23}=0.164$ to 0.25) for the elements in question, measured and assumed in radioactive decay mode in the 1960's, are obviously too large compared with the later semiempirical compilation and theoretical calculations, for example, $f_{23}(\mathrm{Tl})=0.118$ by Krause[1] and $f_{23}(\mathrm{Hg})=0.127$ by Chen *et al.*[35] In the case of intensity measurements of radiative transitions to the L_3 subshell, the overestimates of the f_{23} value are due to overestimates of the number of L_3 vacancies transferred from the L_2 subshell, and may lead to an underestimate of the number of L_3 vacancies transferred from the L_1 subshell and an overestimate of the number of L_2 vacancies from the L_1 subshell. It brings about smaller f_{13} and bigger f_{12} values.[30,32–34,36] Therefore, those earlier data for the L_1 subshell are still questionable, which somewhat misled the semiempirical compilation of Krause.

In 1971, McGuire[37] and Crasemann *et al.*[38] reported, respectively, comprehensive calculations for the L-subshell yields by using different wave functions. Agreement between the two calculations is good.[22] However, exact locations of the discontinuities of the L_1 CK yields as a function of atomic number, where the energy thresholds for some intense groups of L_1 CK transitions are located, are somewhat uncertain.

After that, Chen *et al.* carried out a systematic *ab initio* relativistic calculation of L-subshell CK energies and decay probabilities,[39,40] and then in 1981 reported a new set of decay yields[35] for elements between $Z=18$ and 100. The L_1-subshell results of the new calculation are generally in accord with the above two. From the calculation of CK transition probabilities by Chen *et al.*,[40] we know that L_1-L_2M, L_1-L_2N_1, L_1-$L_3M_{1,2,3}$ and L_2-L_3M CK processes in the considered elements and L_1-$L_3M_{4,5}$ CK processes in Ta and W are all closed, while L_1-$L_3M_{4,5}$ CK processes are energetically allowed in the higher-Z elements. By the way, a number of L_1-subshell CK transition cutoffs and onsets are located in the heavy element region,[39] so that interpolation of the yield values to obtain estimates for neighboring elements must be made with caution.

In the past 15 years, many new measurements of the L_1-vacancy decay yields of heavy elements were reported. However, most of them, e.g., those of Werner and Jitschin[10] and Marques *et al.*,[12] violate the semiempirical compilation and the latest theoretical calculation, which will be discussed in Sec. IV.

B. Origin of the observed satellite lines

The x-ray satellite lines, usually occurring on the high-energy side of the parent diagram lines, arise from radiative transitions in atoms multiply ionized in inner shells or subshells. In high-resolution x-ray and photoelectron spectrometry, some of the satellite lines lie far above their parent line and form an observed satellite structure, while the others fall within the natural width of their parent line and become a part of the observed diagram line.

In the case of intermediate elements Zr and Ag, in the light of relativistic calculations of the energy shift of radiative transitions to the L shell in the presence of an additional vacancy in the M or N shell, Krause *et al.*[41] and Chen *et al.*[17] proposed that the observed satellite lines, shifted at least 7 eV for Zr and from 10 to ~ 30 eV for Ag with respect to their parent L x-ray lines, should be due to LM double-vacancy states created by M-electron shakeoff and shakeup events and L_1-$L_{2,3}M_{4,5}$ CK processes, whereas the satellite lines arising from LN states should be too close to the parent lines to be distinguishable. With this proposal, they interpreted the Zr and the Ag L x-ray spectrum, respectively. Also we have successfully analyzed some L x-ray spectra.[14–16,42]

For heavy elements, in the analyses of the L x-ray spectra of Goldberg,[19] Ferreira *et al.*[20] and Salgueiro *et al.*[21] suggested in 1965 and 1974, respectively, that the observed satellites of $L\alpha$ and $L\beta_2$ lines arise from $L_3M_{4,5}$ doubly ionized atoms produced by L_1-$L_3M_{4,5}$ CK processes, and that the observed satellites of $L\gamma_1$ lines arise from the L_2N_4 ionized atoms created by L_1-L_2N_4 CK processes while the unshifted satellites might be due to L_1-L_2O transitions.

In 1981, Parente *et al.*[43] reported a relativistic computation of L x-ray satellite energies in the presence of one spectator vacancy in the M or N shell for 11 elements in the range $65 \le Z \le 95$, which is consistent with the later theoretical calculations by Uchai *et al.*[44] and by Bhattacharya *et al.*[45] The averaged energy shifts, related to the present work, are listed in Table I.

From Fig. 1 of Ref. 21, we know that the average ener-

TABLE I. Theoretical average energy shifts (in eV) of L x rays in the presence of a spectator vacancy in M or N shell.

		Shell with one spectator vacancy					
Element	Diagram line	M_4	M_5	N_4	N_5	N_6	N_7
W	$L\gamma_1 L_2\text{-}N_4$	63.5	62.7	6.4	6.7	1.0	0.6
	$L\alpha_2 L_3\text{-}M_4$	25.2	23.6	2.9	2.3	−1.5	−1.7
	$L\alpha_1 L_3\text{-}M_5$	25.8	24.0	2.4	1.9	−1.8	−1.6
	$L\beta_2 L_3\text{-}N_5$	65.1	61.9	7.0	5.9	0.7	0.8
Pt	$L\gamma_1 L_2\text{-}N_4$	69.1	67.7	7.6	7.9	1.9	1.4
	$L\alpha_2 L_3\text{-}M_4$	27.0	25.1	3.2	2.5	−1.7	−2.0
	$L\alpha_1 L_3\text{-}M_5$	27.9	25.6	2.6	2.0	−2.3	−2.0
	$L\beta_2 L_3\text{-}N_5$	70.6	66.6	8.3	6.9	1.6	1.6
Hg	$L\gamma_1 L_2\text{-}N_4$	72.0	70.2	8.4	8.6	2.4	1.9
	$L\alpha_2 L_3\text{-}M_4$	28.0	25.9	3.5	2.7	−1.8	−2.1
	$L\alpha_1 L_3\text{-}M_5$	29.0	26.5	2.8	2.1	−2.4	−2.1
	$L\beta_2 L_3\text{-}N_5$	73.4	69.1	9.0	7.6	2.2	2.2

gy shift of the Au $L\gamma_1$ satellite structure with respect to the diagram line is about 60 eV, so we conclude in view of the theoretical data in Table I that the observed satellite structures measured by Salgueiro *et al.*[21] arise from the *LM* doubly ionized atoms produced by simultaneous *LM* ionizations, *M*-electron shakeoff and shakeup events, and L_1-$L_3M_{4,5}$ CK processes, while the satellite lines due to *LN* and *LO* ionized atoms are hidden in the diagram lines.

C. Effect of multiple-vacancy states on L x-ray fluorescence yields

The influence of multiple-vacancy states on the L-subshell decay rates is an important and interesting subject in atomic physics. The multiple-vacancy state of an atom can be produced by simultaneous ionization of atomic shells or subshells by charged particle (particularly heavy ion) bombardments and by shakeoff and shakeup, Auger, and CK events following initial inner-vacancy production. No systematic and judicious theories and measurements on this subject have been worked out hitherto in sufficient detail to give a general numerical result because of the crucial complexity resulting from the multiple-vacancy configurations. Anyway, the presence of a spectator vacancy in some inner shell or subshell brings about a radial contraction of all the electron orbitals of the atom, i.e., a binding-energy increase of all the atomic levels, which causes a change in the energies, intensity ratios, and decay rates of both x-ray and nonradiative transitions. Furthermore, a spectator vacancy can alter the CK rates, which are sensitive to the transition energy, and even causes a certain CK transition to be cut off if it is located near the energy threshold.

Theoretical studies of the fluorescence yields in the case of multiply ionized atoms started in the early 1970s.[46,47] In the meanwhile, some primary measurements were also performed.[47–49] The general conclusion is that both the fluorescence yields and the x-ray intensity ratios of the multiply ionized atoms can be significantly different from those of the singly ionized atoms and vary with the number and configuration of vacancies, which are related to the nature of an excitation process. However, the difference is most important only for the penultimate shell of the atoms when the valence electrons are successively ejected out, but slight for doubly ionized atoms.

In 1977, Rao *et al.*[50] studied Pb radiative decays of the atomic states with *LL*, *LX*, and *MX* double vacancies created by K Auger processes following the electron-capture decay of ^{207}Bi by using coincidence experiments. Next, Campbell *et al.*[51] recorded Ta L x-ray spectra emitted following the radioactive decay of ^{181}W in coincidence with some individual *KLL* Auger electrons selected by a high-resolution magnetic spectrometer. These measurements lead to an inference that the radiative decay yields of L_i-vacancy states, at least for heavy elements, do not seem to be influenced by the presence of a spectator vacancy within the experimental error. For intermediate elements In and Rh, Indira *et al.*[52] and Markevich and Budick[7] also found that the increase in average L x-ray fluorescence yields for double-vacancy states accompanying radioactive decay of ^{113}Sn and ^{103}Pd was small (~10%).

Recently Lorenz and Hartmann[53] calculated, using Dirac-Fock-Slater one-electron wave functions, the effect of an L spectator vacancy on the L x-ray fluorescence yields and relative intensities in a special case of double-vacancy states formed by *KLL* Auger processes. According to that calculation, 1% shifts of transition energies and 10% changes of transition rates for intermediate element In were found; for heavier elements like Pb they were half as large, whereas the calculated changes of both the relative intensities and the average fluorescence yields of L x rays were much less than those measured before. Therefore we can draw from the above information an inference that the influence of an M-shell or an outer-shell spectator vacancy upon the L x-ray fluorescence yields and relative intensities is very small for heavy elements.

D. Electron-ionization cross-section ratios of the three L subshells

The study of atomic inner-shell ionization by impact of electrons began in the 1930s,[54] but both theoretical calculations giving numerical values and systematic measurements are limited, especially for the L subshells.[55,56] In 1961, Victor[57] proposed in accordance with his experiments on electron ionization of the elements in the range $74 \leq Z \leq 92$ that if the ionization was induced by the electrons with energies $E_e > 40$ keV, the relative ionization cross sections of the three L subshells could be calculated, with an error of 15%, from the expression

$$\sigma_1 : \sigma_2 : \sigma_3 :: 1/W_1 : 1/W_2 : 1/W_3 \ , \quad (1)$$

where W_1, W_2, and W_3 denote the electron binding energies of the three L subshells, respectively. Thus the changes of the ratios, σ_2/σ_1 and σ_3/σ_1, are both very small when incident electron energies are more than 40 keV.

In 1978, Scofield[23] presented a relativistic calculation of the L-subshell ionization cross sections by high-energy electrons with incident energies from 50 keV to 1 GeV for the elements Ar, Ni, Y, Ag, Ba, Ho, Au, Bi, and U. The calculations used the first-order Born approximation to deal with the interaction between the scattered and atomic electrons. The high-energy electron was described by plane-wave solutions of the Dirac equation while the atomic electrons were treated relativistically as moving in a Hartree-Slater central potential. The calculation has been tested by a recent measurement of Reusch *et al.*,[56] using a flat crystal spectrometer to measure the characteristic L x rays induced by electrons with energies between 50 and 200 keV, and the comparison between them displays a good agreement for intermediate and heavy elements. The results of both the calculation and the measurement show that the change of cross-section ratios σ_2/σ_1 and σ_3/σ_1 with increasing atomic number is smooth and slight. The theoretical ratios, some of which are interpolated, are given in Table II for the electron energy $E_e = \sim 50$ keV and for the investigated elements. For a comparison, also listed in this table are the values used by Ferreira *et al.*[20] and Salgueiro *et al.*,[21] which are calculated from Eq. (1).

TABLE II. Ionization cross-section ratios by electrons with energy ~50 keV, used in Refs. 20, 21, and in the present work.

	Refs. 20 and 21		Present work	
Element	σ_3/σ_1	σ_2/σ_1	σ_3/σ_1	σ_2/σ_1
Ta	2.36		3.27	1.308
W	2.36	1.048	3.28	1.304
Re		1.048	3.30	1.298
Ir	2.39	1.046	3.34	1.286
Pt	2.40		3.36	1.280
Au	2.41	1.045	3.38	1.272
Hg			3.40	1.264
Tl	2.43		3.42	1.256
Pb			3.44	1.247
Bi			3.47	1.238

III. PRINCIPLE OF THE CALCULATION MODEL

When atomic L subshells are ionized, several inner-vacancy processes, i.e., outer-electron shakeoff (including shakeup), CK, Auger, and x-ray transitions, can take place. L x-ray satellite lines arise from multiply ionized atoms with one vacancy in the L shell, which are produced by the simultaneous inner-shell ionization and concomitant shakeoff events and by the L-shell CK transitions. The observed satellite lines recorded by the high-resolution spectrometer arise from LM double- and LMX $(X = M,N,O,P)$ triple-vacancy states while the satellite lines originating from LN double- and LNN $(N = N,O,P)$ triple-vacancy states are not resolved from their diagram lines.

In order to separate different contributions from the LM and LMX states and the LN and LNN states, the simultaneous ionization and shakeoff probabilities and the CK transition yields are expressed as

$$P_i = P_{iM} + P_{iN} \quad (2)$$

and

$$f_{ij} = f_{ijM} + f_{ijN} \ , \quad (3)$$

where the i subscript denotes the L_1, L_2, and L_3 subshell, P_{ix} is the probability to form L_iX states by the simultaneous ionization and shakeoff events, and f_{ijx} is the L_i-L_jX CK yield. Then the cross sections of emitting true L_i-subshell diagram lines arising from single-vacancy states are given by

$$\sigma(L_1) = \sigma_1(1-P_1)\omega_1 \ , \quad (4)$$

$$\sigma(L_2) = \sigma_2(1-P_2)\omega_2 \ , \quad (5)$$

$$\sigma(L_3) = \sigma_3(1-P_3)\omega_3 \ . \quad (6)$$

The cross sections of emitting L_i diagram lines arising from L_iN double- and L_iNN triple-vacancy states are given by

$$\sigma(L_1N) = \sigma_1 P_{1N} \omega'_{1N} \ , \quad (7)$$

$$\sigma(L_2N) = \sigma_1(1-P_1) f_{12N} \omega'_{2N} + \sigma_2 P_{2N} \omega'_{2N} \ , \quad (8)$$

$$\sigma(L_3N) = \sigma_1(1-P_1) f_{13N} \omega'_{3N} + \sigma_2(1-P_2) f_{23N} \omega'_{3N} + \sigma_3 P_{3N} \omega'_{3N} \ ; \quad (9)$$

$$\sigma(L_1NN) = 0 \ , \quad (10)$$

$$\sigma(L_2NN) = \sigma_1 P_{1N} f_{12N} \omega''_{2NN} \ , \quad (11)$$

$$\sigma(L_3NN) = \sigma_1 P_{1N} f_{13N} \omega''_{3NN} + \sigma_1(1-P_1) f_{12N} f_{23N} \omega''_{3NN} + \sigma_2 P_{2N} f_{23N} \omega''_{3NN} \ . \quad (12)$$

The cross sections of emitting the observed satellite lines due to L_iM double- and L_iMX triple-vacancy states are given by

$$\sigma(L_1M)=\sigma_1P_{1M}\omega'_{1M} , \tag{13}$$

$$\sigma(L_2M)=\sigma_1(1-P_1)f_{12M}\omega'_{2M}+\sigma_2P_{2M}\omega'_{2M} , \tag{14}$$

$$\sigma(L_3M)=\sigma_1(1-P_1)f_{13M}\omega'_{3M}+\sigma_2(1-P_2)f_{23M}\omega'_{3M}+\sigma_3P_{3M}\omega'_{3M} ; \tag{15}$$

$$\sigma(L_1MX)=0 , \tag{16}$$

$$\sigma(L_2MX)=\sigma_1P_{1N}f_{12M}\omega''_{2MN}+\sigma_1P_{1M}f_{12N}\omega''_{2MN}+\sigma_1P_{1M}f_{12M}\omega''_{2MM} , \tag{17}$$

$$\begin{aligned}\sigma(L_3MX)=&\sigma_1(P_{1M}f_{13N}\omega''_{3MN}+P_{1N}f_{13M}\omega''_{3MN}+P_{1M}f_{13M}\omega''_{3MM})\\&+\sigma_1(1-P_1)(f_{12N}f_{23M}\omega''_{3MN}+f_{12M}f_{23N}\omega''_{3MN}+f_{12M}f_{23M}\omega''_{3MM})\\&+\sigma_2(P_{2M}f_{23N}\omega''_{3MN}+P_{2N}f_{23M}\omega''_{3MN}+P_{2M}f_{23M}\omega''_{3MM}) ,\end{aligned} \tag{18}$$

where σ_i is the ionization cross section, and ω'_{ix} and ω''_{ixy} are the L_i-subshell fluorescence yields modified due to the L_iX- and L_iXY-vacancy states, respectively. We set

$$\omega'_{ix}=q_1\omega_i , \tag{19}$$

$$\omega''_{ixy}=q_2\omega_i , \tag{20}$$

where q_1 and q_2 are the parameters referred to the influence of one and two spectator vacancies on the L_i-subshell fluorescence yield ω_i, respectively, and $q_1\approx q_2\approx 1$ for heavy elements for the reason described in Sec. II. Here, it is assumed in Eqs. (13)–(18) that the L_i-subshell vacancies in the double- and triple-vacancy states always decay before the radiative and Auger transitions of the outer M vacancies, which have been populated in $M_{4,5}$ subshells due to the intense M-shell CK processes, i.e., the $M_{4,5}$ vacancies remain as the spectators while the L_i vacancies are being filled by radiative transitions.

For the heavy elements investigated, partial CK yields $f_{12M}=0$ and $f_{23M}=0$, as mentioned above. According to Slater's recipe,[58] the shakeoff probabilities due to one L_1, L_2, or L_3 vacancy are approximately equal, so $P_{1M}=P_{2M}=P_{3M}=P_M$, $P_{1N}=P_{2N}=P_{3N}=P_N$, $P_1=P_2=P_3=P$, and $P=P_M+P_N$. Therefore the cross sections of emitting L_1-, L_2-, and L_3-subshell diagram, satellite, and total lines are given by

$$\sigma^d_{L_1}=(1-P+P_N)\sigma_1\omega_1 , \tag{21}$$

$$\sigma^s_{L_1}=P_M\sigma_1\omega_1 ; \tag{22}$$

$$\sigma^d_{L_2}=(1-P+P_N)(\sigma_2+\sigma_1f_{12})\omega_2 , \tag{23}$$

$$\sigma^s_{L_2}=P_M(\sigma_2+\sigma_1f_{12})\omega_2 ; \tag{24}$$

$$\begin{aligned}\sigma^d_{L_3}=&(1-P+P_N)(\sigma_3+\sigma_2f_{23}+\sigma_1f_{13N})\omega_3\\&+(1-P)\sigma_1f_{12}f_{23}\omega_3 ,\end{aligned} \tag{25}$$

$$\sigma^s_{L_3}=P_M(\sigma_3+\sigma_2f_{23}+\sigma_1f_{13N})\omega_3+\sigma_1f_{13M}\omega_3 ; \tag{26}$$

$$\sigma_{L_1}=\sigma^d_{L_1}+\sigma^s_{L_1}=\sigma_1\omega_1 , \tag{27}$$

$$\sigma_{L_2}=\sigma^d_{L_2}+\sigma^s_{L_2}=(\sigma_2+\sigma_1f_{12})\omega_2 , \tag{28}$$

$$\sigma_{L_3}=\sigma^d_{L_3}+\sigma^s_{L_3}=(\sigma_3+\sigma_2f_{23}+\sigma_1f_{13})\omega_3 , \tag{29}$$

where the d and s superscripts denote the observed diagram and satellite x-ray lines, respectively. If we set $P_M=0$ in Eqs. (22) and (24), $\sigma^s_{L_1}=\sigma^s_{L_2}=0$. Hence the observed satellite lines of the radiative transitions to the L_1 and L_2 subshells arise from simultaneous LM ionization and M-electron shakeoff events. Equation (26) states that the observed satellite lines of the radiative transitions to the L_3 subshells are made up from two different processes: one is simultaneous LM ionization and M-electron shakeoff events, and the other is L_1-$L_3M_{4,5}$ CK transitions if allowed. Using the x-ray intensity ratio $I^s_{L_i}/I^d_{L_i}$ to take the place of the ratio of $\sigma^s_{L_i}/\sigma^d_{L_i}$ and neglecting the small contribution of the last term in Eq. (25), we have the following expressions:

$$I^s_{L_1}/I^d_{L_1}=I^s_{L_2}/I^d_{L_2}=P_M/(1-P+P_N) , \tag{30}$$

$$\begin{aligned}I^s_{L_3}/I^d_{L_3}=&P_M/(1-P+P_N)\\&+(I_{L_3}/I^d_{L_3})[\sigma_1f_{13M}/(\sigma_3+\sigma_2f_{23}+\sigma_1f_{13})] .\end{aligned} \tag{31}$$

Therefore, if the L_1-L_3M CK transitions in the investigated elements are closed,

$$(I^s_{L_3}/I^d_{L_3})_{f_{13M}=0}=I^s_{L_1}/I^d_{L_1}=I^s_{L_2}/I^d_{L_2} , \tag{32}$$

and vice versa. Then the probability P_M, partial CK transition yield f_{13M}, and L_1-subshell fluorescence yield ω_1 are given by

$$P_M=(I^s/I)_{L_1}=(I^s/I)_{L_2} , \tag{33}$$

$$\begin{aligned}f_{13M}=&(\sigma_3/\sigma_1+\sigma_2f_{23}/\sigma_1+f_{13})(I^d/I)_{L_3}\\&\times[(I^s/I^d)_{L_3}-(I^s/I^d)_{L_2}] ,\end{aligned} \tag{34}$$

$$\omega_1=(\sigma_2/\sigma_1+f_{12})(I_{L_1}/I_{L_2})\omega_2 , \tag{35}$$

$$\omega_1=(\sigma_3/\sigma_1+\sigma_2f_{23}/\sigma_1+f_{13})(I_{L_1}/I_{L_3})\omega_3 . \tag{36}$$

In Eqs. (35) and (36), the modification factors (q_1 and q_2, if written into) and CK yields (f_{12}, f_{13}, and f_{23}) appear in the terms that are much smaller than the others in the related sums, so that both their uncertainties and the modifications of the CK yields due to double- and triple-vacancy states are unimportant in present calculations of the ω_1.

Equations (35) and (36) provide a simple approach for calculating ω_1 values from some known data of ω_2 and ω_3 belonging in the same L shell. In addition to L x-ray spectra recorded by high-resolution crystal spectrometers, this new approach may also be applicable to some spectra, of heavy elements, recorded by Si(Li) detectors if the peak-deconvolution technique adopted is excellent.

The two equations can be easily derived also from the following well-known expressions for singly ionized atoms:

$$I_{L_1}=n_1\omega_1 \ , \tag{37}$$

$$I_{L_2}=n_2\omega_2+n_1 f_{12}\omega_2 \ , \tag{38}$$

$$I_{L_3}=n_3\omega_3+n_2 f_{23}\omega_3+n_1(f_{13}+f_{12}f_{23})\omega_3 \ , \tag{39}$$

if we use ionization cross-section ratios σ_2/σ_1 and σ_3/σ_1 to take the place of initial vacancy ratios n_2/n_1 and n_3/n_1, respectively, and neglect the small contribution from the term of $n_1 f_{12} f_{23}\omega_3$ in Eq. (39). Hence the physical meaning of Eqs. (35) and (36) is very clear.

IV. RESULTS AND DISCUSSION

Goldberg[19] reported in 1962 the relative intensities of L x-ray lines for the heavy elements, which were excited by electrons in an x-ray tube worked at the voltage ~50 kV and the current ~25 mA. The measurements were made with a curved mica crystal bent to a cylinder of 40 cm radius. The anode was composed of a copper supporter of an element target. The electrodeposited metallic coat of several micrometers thick was used for elements Pt, Au, Hg, Tl, Pb, Bi, and Re (75 at. % Re+25 at. % Ni). The Ta and W were covered with copper and then soldered on the supporter. The thickness of the deposited coat for Ir is about 1 μm.

Ferreira *et al.*[20] evaluated in 1965 the intensity ratios of the $L\alpha$ and the $L\beta_2$ satellite to total lines, I^s_α/I_α and $I^s_{\beta_2}/I_{\beta_2}$, using a spectrometer with a curved quartz crystal of 35 cm radius, while Salgueiro *et al.*[21] estimated in 1974 the relative intensity of the $L\gamma_1$ satellite and total line, $I^s_{\gamma_1}/I_{\gamma_1}$, induced by 42-keV electrons with a curved quartz crystal of 20 radius. Goldberg's results related to the present work are listed in Table III but some of them are recalculated according to the data given in Refs. 20 and 21. Also the relative intensities of the observed satellite and diagram lines for the $L\gamma_1$ and $L\beta_2$ lines are listed in Table III.

In the present calculations, Krause's CK yields f_{12}, f_{13}, and f_{23} (Ref. 1) and Scofield's theoretical electron-ionization cross sections given in Table II are adopted.

First, using Goldberg's relative intensities of the L x-ray lines, we calculated from Eqs. (35) and (36) the ratios of the fluorescence yields ω_1/ω_2, ω_1/ω_3 as well as ω_2/ω_3. They are given in Table IV, along with some recent measurements. This table shows that though some systematic differences exist, the present results are generally in good agreement with the latest ones of Werner and Jitschin[10,11] and Campbell and McGhee[59] within the listed errors except the value of ω_1/ω_3(Ta)=0.757, which is too large as pointed out in Ref. 10. It is a primary test of Goldberg's data. However, the present results do not favor those of Kodre *et al.*,[60] Tan *et al.*,[8] and Rao,[61] and also apparently those of the semiempirical compilation[1] and the theoretical calculation[35] for the elements with $Z\approx 80$. Then we tried to obtain reasonable values of the L_1-subshell fluorescence yields ω_1. In Eqs. (35) and (36), both the ionization cross sections and the x-ray intensities appear as ratios, which reduces appreciably systematic errors, but the uncertainties of adopted values of ω_2 and ω_3 can directly transfer to resulting ω_1 values.

Recently Campbell and McGhee[59] have made an x-ray coincidence measurement of ω_2 and ω_3 for elements Au, Hg, Tl, and Pb by using radioisotopes ^{195}Au, ^{203}Hg, ^{204}Tl, and ^{207}Bi. Their values agree well with the latest theoretical data[35] to within a typical error of 2–3 % but do not favor the semiempirical compilation, which tends to lie about 7.5% below Krause's data for ω_3 and about 6% above Krause's data for ω_2. Werner and Jitschin[10] have adopted 0.96 times Krause's data as the best estimates of ω_3 for the elements with $72\le Z\le 82$ investigated in their work, and calculated ω_2 and ω_1 values in the light of the

TABLE III. Relative intensities of the L_1-, L_2-, and L_3-subshell x rays and intensity ratios of the satellite, diagram, and total x rays for $L\gamma_1$ and $L\beta_2$ lines (Refs. 19–21).

Element	I_{L_1}	I_{L_2}	I_{L_3}	$I^s_{\gamma_1}/I_{\gamma_1}$	$I^s_{\gamma_1}/I^d_{\gamma_1}$	$I^s_{\beta_2}/I_{\beta_2}$	$I^s_{\beta_2}/I^d_{\beta_2}$
Ta	21.5	57.0	135.3			0.046±0.005	0.048
W	18.17	60.1	135.9	0.099	0.110	0.05±0.004	0.05
Re	15.3	58.6	136	0.105	0.117		
Ir	18.8	69.4	136.0	0.063	0.067	0.09±0.008	0.10
Pt	13.8	62.5	136.1			0.10±0.01	0.11
Au	14.1	59.4	137.6	0.080	0.087	0.11±0.01	0.12
Hg	13.7	61.1	135				
Tl	13.9	63.7	137.2			0.11±0.02	0.12
Pb	13.5	56.7	138				
Bi	12.8	55.3	138				
Uncertainty	10%	9%	2%	15%			

TABLE IV. Ratios of L-subshell fluorescence yields, ω_1/ω_2, ω_1/ω_3, and ω_2/ω_3. The data in parentheses are not in line with the others related.

	ω_1/ω_2			ω_1/ω_3			ω_2/ω_3				
Element	Present	Werner and Jitschin (Ref. 10)	Others	Present	Werner and Jitschin (Ref. 10)	Others	Present	Werner and Jitschin (Ref. 10)	Campbell and McGhee (Ref. 59)	Others	Adopted
$_{72}$Hf		0.511			0.561			1.098			
$_{73}$Ta	0.561			0.592	(0.757)		1.06	1.127			1.08
$_{74}$W	0.446	0.472		0.499	0.530		1.12	1.123			1.12
$_{75}$Re	0.381			0.427			1.12				1.14
$_{77}$Ir	0.389	0.430		(0.546)	0.494			1.148		(1.02)[a]	1.18
$_{78}$Pt	0.314	0.372		0.408	0.443		1.30	1.189			1.20
$_{79}$Au	0.335	0.346	0.337[b]	0.417	0.446	0.422[b]	1.24	(1.289)	1.20	1.25[b]	1.20
$_{80}$Hg	0.313			0.417			1.33		1.19		1.20
$_{81}$Tl	0.302			0.419					1.22		1.20
$_{82}$Pb	0.326	0.356	(0.393)[c] 0.284[d]	0.407	0.420	(0.461)[c] (0.344)[d]	1.25	1.180	1.17	1.17[c] 1.21[d]	1.19
$_{83}$Bi	0.312			0.389			1.24				1.18
Error	14%	7%		11%	6%		18%	4%	3.3%		

[a]From Rao (Ref. 61).
[b]From Jitschin *et al.* (Ref. 11).
[c]From Kodre *et al.* (Ref. 60).
[d]From Tan *et al.* (Ref. 8).

ratios ω_2/ω_3 and ω_1/ω_3 directly measured by employing the synchrotron photoionization method. In general, these ω_2 and ω_3 values are in accordance with Krause's compilation for the elements Hf, Ta, and W, and with the latest theoretical calculation for higher-Z elements Ir, Pt, Au, and Pb.

In order to acquire a proper group of ω_2 and ω_3 values for the elements under study, we have calculated four kinds of ω_1 values by using the ω_2 and ω_3 data taken from the semiempirical compilation of Krause, theoretical calculation (interpolated) of Chen *et al.*, direct measurements of Campbell and McGhee,[59] and analytical estimates of Werner and Jitschin,[10] respectively, and two groups from Eqs. (35) and (36) for each kind. The calculated values show that for the elements Ta, W, and Re the agreement between the two groups of ω_1 values in the three cases (no data in the case of Campbell and McGhee) are all very good, while for the elements between $Z=77$ and 83 the last two are the best of the four cases and Krause's data gives disagreement.

From analyses of these calculated ω_1 values as well as the recent measurements of ω_2 and ω_3,[10,59–63] we suggest the ω_3 values given in Table V as the best estimates in this work, which generally follow the compiled data and the estimates of Werner and Jitschin at $Z=73$ and 74, and follow the measured values of Campbell and McGhee for $Z \geq 79$. The corresponding ω_2 values are evaluated by balancing the two groups of ω_1 values calculated from Eqs. (35) and (36) and also given in Table V. The practically adopted ratios ω_2/ω_3 are presented in the last column of Table IV. The errors of the ω_2 and ω_3 values, given at the bottom of Table V, are estimated. We find that the present estimates of ω_2 are in line with those of Werner and Jitschin.[10] The present ω_3 and ω_2 values are also in fair agreement with all other recent measurements[8,60,61,63] to within the given uncertainties, though the values of Tan *et al.*[8] are systematically smaller.

Two kinds of ω_1 values have been obtained by using the ω_2 and ω_3 values taken, respectively, from the present estimates and from the latest theory. The results are given in Table V and Fig. 1, which are averages of the two groups of values. The results show that the two kinds of ω_1 values are quite consistent though the latter are a little larger than the former for the lighter elements under consideration, which is due to the somewhat larger ω_2 and ω_3 values from the theory.

All experimental data of ω_1 available from literature, along with the present ones, are given in Fig. 1. The compiled and the theoretical data are also plotted for a comparison. Figure 1 shows that the present two kinds of ω_1 values are in agreement with those measured after

TABLE V. The estimated L_3 and L_2 fluorescence yields ω_3 and ω_2, and the calculated L_1 fluorescence yields ω_1. In this table, Est. and Theor. indicate that the ω_1 values are calculated by adopting the estimated and the theoretical values of ω_3 and ω_2, respectively. The errors listed at the bottom are estimated.

			ω_1	
Element	ω_3	ω_2	Est.	Theor.
$_{73}$Ta	0.241	0.260	0.144	0.154
$_{74}$W	0.246	0.275	0.123	0.131
$_{75}$Re	0.257	0.293	0.111	0.116
$_{77}$Ir	0.274	0.323	0.126	0.128
$_{78}$Pt	0.284	0.341	0.112	0.115
$_{79}$Au	0.296	0.355	0.121	0.124
$_{80}$Hg	0.310	0.372	0.123	0.124
$_{81}$Tl	0.324	0.389	0.127	0.127
$_{82}$Pb	0.340	0.405	0.135	0.134
$_{83}$Bi	0.354	0.418	0.134	0.132
Error	4%	5%	10%	10%

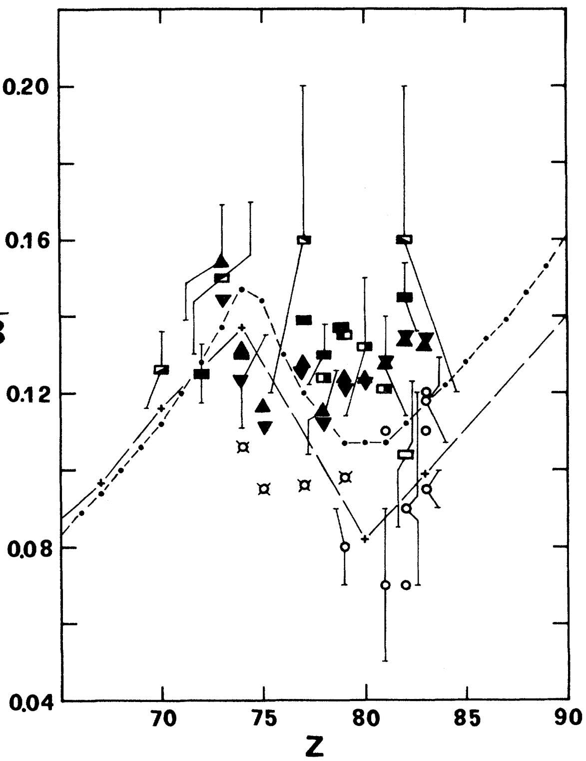

FIG. 1. L_1-subshell fluorescence yields ω_1 as a function of atomic number Z. Small dots and crosses are the compiled (Ref. 1) and the theoretical data (Ref. 35), and connected by lines to guide the eye, respectively. Experimental data: ▲ and ▼, obtained in present work by using the theoretical and the estimated ω_2 and ω_3 values, respectively; ○ and ¤, reported before 1975 (collected in Refs. 1, 12, 21, 22, 25, and 30), the latter with antennae denotes the values of Salgueiro *et al.* (Ref. 21); ■, Werner and Jitschin (Ref. 10); ■, Jitschin *et al.* (Ref. 11); ■, Marques *et al.* (Ref. 12); □, Tan *et al.* (Ref. 8); ■, McNelles *et al.* (Ref. 66); ■, Indira *et al.* (Ref. 64); ■, Indira *et al.* (Ref. 65); ■, Kodre *et al.* (Ref. 60). Some representative error bars are plotted.

1975, particularly with the latest ones of Werner and Jitschin[10,11] and of Marques *et al.*,[12] that all the new experimental values, except that of Tan *et al.*,[8] are much larger than the old ones measured before 1975 (including those of Salgueiro *et al.*[21]), the latter are unreasonable as indicated in Sec. II, and that these latest[10–12] and present values do not change much with increasing atomic number and are not in agreement with the semiempirical compilation and the theoretical calculation for the elements $Z \approx 80$. In general, they are in line with the compilation and the theory for the elements around $Z = 73$, but $\sim 20\%$ more than the compiled ones and $\sim 55\%$ more than the theoretical ones at $Z \approx 80$. Thus the latest theory overestimates appreciably the L_1-subshell CK rates at $Z \approx 80$.

In addition, assuming

$$I^s_{L_2}/I^d_{L_2} = I^s_{\gamma_1}/I^d_{\gamma_1}, \tag{40}$$

$$I^s_{L_3}/I^d_{L_3} = I^s_{\beta_2}/I^d_{\beta_2}, \tag{41}$$

we found the values of P_M and f_{13M} from Eqs. (33) and (34) and the experimental intensity ratios listed in Table III. The present L_1-L_3M CK yields $f_{13M}(\mathrm{Ir}) = 0.115 \pm 0.050$ and $f_{13M}(\mathrm{Au}) = 0.133 \pm 0.055$. Werner and Jitschin[10] and Indira *et al.*[64] reported the measured value of $f_{13}(\mathrm{Ta,W}) = f_{13N} \approx 0.33$, so the estimates of the L_1-L_3 CK yields for Ir and Au are $f_{13}(\mathrm{Ir}) = 0.45$ and $f_{13}(\mathrm{Au}) = 0.46$. Compared with the measurement of Werner and Jitschin,[10] the estimate for Au is considerably smaller, which indicates that the old technique used in Refs. 20 and 21 is somewhat questionable for peak deconvolution of the satellite and diagram x-ray spectra. However, the ratios $I^s_{\beta_2}/I^d_{\beta_2}$ and $I^s_{\gamma_1}/I^d_{\gamma_1}$ display clearly that the L_1-$L_3M_{4,5}$ CK transitions are forbidden in Ta and W but allowed in Ir, Pt, etc. On the other hand, the appreciable decrease of the ω_1 values at $Z = 75$ and 78 (see Fig. 1) indicates that the onset of L_1-L_3M_5 CK transition is located at $Z = 75$ and the onset of L_1-L_3M_4 transition at $Z = 78$, which are consistent with the theoretical prediction of L-subshell CK energy calculation made by Chen *et al.*[39]

By the way, besides the experimental errors and the multiple-hole configurations described in Sec. II, variation of the physical and chemical environment of an atom influences appreciably values of the inner-vacancy decay yields, particularly for the elements located in CK transition onset or cutoff regions,[16] which the elements investigated in this work are just in. However, the influence upon the decay yields of L vacancies for heavy elements is much less than that for light and intermediate elements.

V. CONCLUSIONS

From the information concerning the energy shift of the satellite lines, it was concluded that the observed satellite lines of the heavy elements under consideration arise from the LM doubly and LMX triply ionized atoms produced by simultaneous LM ionization, M-electron shakeoff and shakeup events and L_1-$L_3M_{4,5}$ CK processes while the satellite lines due to the LN and LNN ionized atoms are not resolved from the parent lines. Then we calculated the L-subshell fluorescence yields of ten elements with $Z = 73$ to 83 from the new model that takes into account the double- and triple-vacancy states with one vacancy in the L shell and the role of possible modifications of the L-vacancy decay yields in the states.

This work gives some interesting results for the elements under consideration. (1) The obtained values of the L-subshell fluorescence yields confirm recent measurements. (2) The old values of ω_1 reported before 1975 are all too small. (3) The semiempirical compilation of ω_1, ω_2, and ω_3 succeeds for the elements with $Z \approx 73$ but fails for the elements around $Z \approx 80$. (4) The change of ω_1 with increasing atomic number in this region of the periodic table is much less than that predicted by the theory and by the compilation. (5) The new experimental values of ω_1 are approximately 20% larger than the compiled data and 55% larger than the latest theoretical predictions for the elements with $Z \approx 80$. (6) The onsets of L_1-L_3M_5 and L_1-L_3M_4 CK transitions are probably located at $Z = 75$ and 78, respectively.

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