L_1- and L_2-subshell fluorescence yields of lanthanides

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By analyzing the L-x-ray spectra induced by 2-MeV proton impact, the L_1- and L_2-subshell fluorescence yields of La, Nd, Dy, Yb, and Lu have been derived. The present yields are in good agreement with recent calculations employing Dirac-Hartree-Slater wave functions, the semiempirical compilation, and also recent measurements, within the uncertainties.

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Fluorescence yields are important physical quantities. In 1979, Krause carried out a semiempirical compilation of atomic L_i-subshell x-ray fluorescence yields ω_i ($i=1$, 2, and 3), Auger transition yields a_i, and Coster-Kronig transition yields f_{12}, f_{13}, and f_{23} for the elements with atomic number $Z=12-110$ [1], in which the assessment of the L_1-subshell yields was mainly based on some existing theoretical calculations pertaining to singly ionized atoms because only a few experimental data were available at that time. In 1981, Chen, Crasemann, and Mark performed a new *ab initio* relativistic calculation of the yields for the selected atoms with $18 \le Z \le 100$ by resorting to perturbation theory with the Dirac-Hartree-Slater (DHS) wave function [2]. Both the compilation and the calculation have been widely used. However, many measurements made in the past decade for intermediate ($Z \approx 40-50$) and heavy elements ($Z \ge 73$) tested them and remarkable discrepancies were found [3–8].

For lanthanides, experimental values reported up to now are very spare except the L_2-L_3 Coster-Kronig yield f_{23}. We have noticed that all f_{23} values recently measured are less than the theoretical ones [8–11], which might imply that the theoretical data of the yield ω_2 are somewhat misfitted. As for the L_1 subshell, both the L_1-L_2 and L_1-L_3 Coster-Kronig yields, f_{12} and f_{13}, as a function of atomic number, change smoothly in the

TABLE I. Relative L-x-ray intensities (normalized to $I_{\alpha_{1,2}}=100$) for 2-MeV proton impact on thin Au targets.

		Experiment				
Line	Present calculation	Cohen [21]	Tawara *et al.* [22]	Sokhi and Crumpton [22]	Sarkadi and Mukoyama [22]	Akselsson and Johansson [23]
L_l	5.05	4.87	5.43	4.90		4.93
L_α	100	100	100	100	100	100
L_η	0.84	1.27[a]	0.65			
$L_{\beta_{1,2,3}}$	49.74	52.7				
$L_{\beta_{4,6}}$	2.42	3.16				
L_{β_5}	1.99	2.04				
L_{γ_1}	6.39	6.38			6.62	
$L_{\gamma_{2,3,6}}$	1.36	1.38			1.48	
$L_{\gamma_{4,4'}}$	0.12	0.12				
L_{γ_5}	0.22	0.35[a]				
L_β	56.4	58.7	56.3	56.3	59.6	58.0
L_γ	8.14	8.50	8.49	7.60	8.74	7.78
L_{total}	170.4	174.7	170.8	168.8	174.2	170.6

[a]Data with large errors due to background removal and low counting statistics in the experiment (see Ref. [21]).

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lanthanide regime and various theoretical values are all better in line with each other [2,12]. Nevertheless, just recently Stötzel *et al.* [11,13] studied experimentally by means of the monochromatized synchrotron-radiation photoionization method the decay channels of Sm L-subshell vacancies and obtained the value of $f_{13}=0.18\pm0.03$ [11] ($f_{13}=0.19\pm0.03$ [13]), which is surprisingly much smaller than all reported data, compiled with the semiempirical approach ($f_{13}=0.30$), predicted by different theories and extrapolated from the experimental values for its neighboring elements [14,15]. The yield f_{13} is an important parameter that may affect the ω_1 value. Hence a further investigation for lanthanides is needed. Present yield values will be acquired from the relative L-x-ray intensities induced by 2-MeV proton impact.

The L_i-subshell fluorescence yields ω_i are related to the L_i-subshell ionization cross section σ_i, x-ray intensities I_i, and Coster-Kronig yields f_{ij} as follows [7]:

$$\omega_1=(\sigma_3/\sigma_1+f_{23}\sigma_2/\sigma_1+f_{13})(I_1/I_3)\omega_3 ,$$

$$\omega_2=(\sigma_3/\sigma_1+f_{23}\sigma_2/\sigma_1+f_{13})(\sigma_2/\sigma_1+f_{12})^{-1}(I_2/I_3)\omega_3 .$$

For the present work, the Coster-Kronig yields all lie in the small terms in the related sums in the above expressions and their exact values are less important. In the following computations, we adopted the compiled f_{ij} values of Krause [1]. The ratio form of the L_i-subshell x-ray intensities and ionization cross sections is beneficial to reduction of systematic errors of the results calculated in this work.

In this work, the ECPSSR ionization cross sections [16] were used, which were calculated in the plane-wave Born approximation (PWBA) with corrections for energy loss (E), Coulomb deflection (C), perturbed stationary state (PSS), and relativistic effects (R). The ECPSSR L-subshell cross sections are justified at least for a few-MeV proton incident on heavy elements [17,18]. As a supplement to Ref. [17], we computed even relative intensities of the individual L-x-ray lines induced by 2-MeV proton bombardment on gold, by using the tabulated ECPSSR cross sections [19], the compiled Coster-Kronig yields, the Dirac-Fock (DF) x-ray emission rates [20], and the fluorescence yields presented by us [7]. The calculated results are given in Table I, together with the measurements of Cohen [21] and others [22,23]. Agreement between them is found to be excellent for all listed lines except for the weak L_η and L_{γ_5}. The two lines, which are located, respectively, between the intense L_α and L_β lines and between the intense L_β and L_{γ_1} lines in Au L-x-ray spectrum, were measured with large errors [21].

The relative L-x-ray intensities measured by Hirokawa, Nishiyama, and Kiso [24] will be used to acquire ω_1 and ω_2 values. The individual lines, induced by proton impact on thin lanthanide-chloride targets and recorded with a high-purity Ge-detector spectrometer, are L_l, L_η, L_{β_1}, $L_{\beta_{2,15}}$, L_{β_3}-L_{β_6}, $L_{\beta_{9,10}}$, L_{γ_1}-L_{γ_6} for Nd, Dy, and Yb and L_l, L_η, L_{β_1}-L_{β_4}, L_{β_6}, $L_{\beta_{15}}$, L_{γ_1}-L_{γ_3} for La and Lu. In order to correct the quantities affecting the relative intensities, empirical factors were applied and their values were determined by the least-squares method in the measurements. The measured relative intensities of the L_i-M and L_i-N transition groups for proton energy $E=2$ MeV are listed in Tables II and III. In Table III, the values for the weak lines L_{β_5}, $L_{\beta_{9,10}}$, L_{γ_4}, and L_{γ_5} are added, which are evaluated by using the relativistic Hartree-Slater (HS) x-ray emission rates [25]. The values in the two tables are normalized to the intensities of the $L_{\alpha_{1,2}}$ and the L_{α_1} lines, respectively.

The computed results are given in Table IV, with estimated uncertainties of 12% for ω_1 and 10% for ω_2, and plotted in Figs. 1 and 2 together with some experimental data published previously [4,5,7–11,14,15,26–30]. In this computation the well-known values of the compiled ω_3 [1] were adopted to derive the less-well-known ω_1 and ω_2 values. The latest theoretical and compiled data are also given in the two figures. Figure 1 shows that the present ω_2 values are located between the compiled and the latest theoretical data and are in good agreement with the reported measurements. The experimental value of Stötzel *et al.* is acquired from a synchrotron-radiation ionization mode [11] and the others are from nuclear

TABLE II. Relative L-x-ray intensities (normalized to $I_{\alpha_{1,2}}=100$) measured for Nd, Dy, and Yb at proton energy $E=2$ MeV.

Transition	Nd	Dy	Yb
L_1-M	12.61	10.20	8.07
L_1-N	3.69	2.75	2.23
L_2-M	41.79	38.72	37.42
L_2-N	7.50	7.44	7.23
L_3-M	103.98	104.23	104.43
L_3-N	19.00	19.09	18.99

TABLE III. Relative L-x-ray intensities (normalized to $I_{\alpha_1}=100$) for La and Lu at proton energy $E=2$ MeV. The standard deviations are given in parentheses.

Transition	La	Lu
L_1-M	16.77(0.31)	7.88(0.63)
L_1-N	5.00(0.02)	2.31(0.11)
L_2-M	47.46(0.46)	41.95(0.89)
L_2-N	8.64(0.05)	8.16(0.18)
L_3-M	115.63	116.31
L_3-N	21.04(0.28)	21.15(0.26)

TABLE IV. L_1 and L_2 fluorescence yields.

Element	ω_1	ω_2
La	0.0642	0.110
Nd	0.0728	0.134
Dy	0.109	0.190
Yb	0.136	0.240
Lu	0.135	0.258

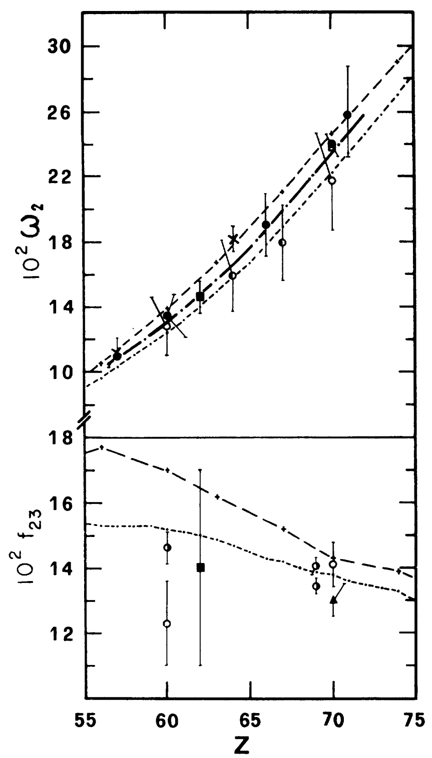

FIG. 1. L_2-subshell fluorescence yield ω_2 and L_2-L_3 Coster-Kronig yield f_{23} as a function of atomic number Z. Small dots and crosses are, respectively, the compiled [1] and the latest [2] theoretical data, connected by lines to guide the eye. Experimental data: ■, Stötzel *et al.* [11]; ○, Tan *et al.* [10]; ◐, Gnade *et al.* [26]; ◑, Catz *et al.* [9]; ▲, McGhee and Campbell [8]; ×, Douglas [27]; □, McNelles *et al.* [15]; ●, present values with typical error bars. The long-dash–dotted line displays the recommended ω_2 values (see text).

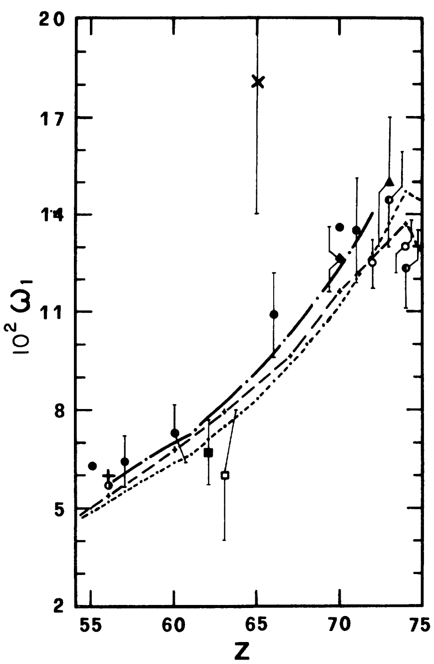

FIG. 2. L_1-subshell fluorescence yield ω_1 as a function of atomic number Z. Small dots and crosses are, respectively, the compiled [1] and the latest [2] theoretical data, connected by lines to guide the eye. Experimental data: ■, Stötzel *et al.* [11]; ◑, Xu and Rosato [4]; +, Burford and Haynes [28]; □, Veluri and Rao [14]; ×, McGeorge *et al.* [30]; ♦, McNelles *et al.* [15]; ▲, Indira *et al.* [29]; ○, Werner and Jitschin [5]; ◐, Xu [7]; ●, present values with typical error bars. The long-dash–dotted line displays the recommended ω_1 values (see text).

disintegration ones. This figure also shows that the experimental values of the L_2-L_3 Coster-Kronig yields, f_{23}, published in the past several years [8–11] are obviously less than the latest theoretical predictions and slightly lower than the compiled data. This trend of departure has also been observed recently in the case of other intermediate and heavy atoms ($Z \approx 80$), and so the theoretical calculation of f_{23} seems to be somewhat unfavorable. We know from the basic relation $\omega_2 + f_{23} + a_2 = 1$ that the overestimate of the compiled f_{23} values may lead to an underestimate of the ω_2 values. On an average, this situation is displayed in Fig. 1, which was also found in recent work for the elements with $Z \approx 80$ [5,7,8].

Figure 2 shows that the present ω_1 values are all slightly larger than the latest theoretical and compiled data, but in accordance with the compiled data within the uncertainty of 15%, given by Krause [1], and also with all the measurements within the error bars [4,5,7,11,14,15,28,29] except that reported by McGeorge, Freund, and Fink [30]. The last one is obviously questionable.

Additionally, in Fig. 2 the experimental ω_1 values for tungsten ($Z = 74$) are a little less than those for tantalum ($Z = 73$). It hints that the onset of the L_1-L_3M Coster-Kronig transition is located at $Z = 74$ rather than $Z = 75$ predicted by the theories [12,31] and by the compilation [1]. This anomaly was also indicated by Salgueiro, Carvalho, and Parente [32] by virtue of a designed experiment to observe the L_{β_2} satellite induced by electron impact on tungsten.

In brief, from an analysis of the L-x-ray intensity spectra produced by 2-MeV proton impact, the L_1- and L_2-subshell fluorescence yields of the elements La, Nd, Dy, Yb, and Lu have been obtained. Within the uncertainties they agree with the latest relativistic calculations, the semiempirical compilation, and also recent measurements. By observing the experimental data in Figs. 1 and 2, we recommend 1.05 times the compiled ω_2 data and 1.10 times the compiled ω_1 data as more reasonable values of the L_2- and L_1-subshell fluorescence yields (long-dash-dotted lines in the two figures), respectively.

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