Fluorescence Yield for the L_2 Subshell in Curium

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Submitted August 13, 1971

Zh. Eksp. Teor. Fiz. **62**, 31–34 (January, 1972)

The spectrum of the characteristic L radiation produced in α decay of ^{252}Cf is measured with an Si(Li) high-resolution spectrometer. The contributions of the L_2 and L_3 subshells in the spectrum are separated and on this basis the fluorescence yield for the L_2 subshell of the Z=96 atom is determined. The result is compared with predictions of the semiempirical Listengarten theory.

THE mechanism of realignment of the internal electron shells of the atom has recently become the subject of more intense study. A number of theoretical[1-3] and experimental[4-6] studies have been devoted, in particular, to the determination of the rates of radiative transitions. Whereas in the theory this is due both to the general progress in our knowledge of intraatomic interactions and to the greater capabilities of electronic computers, in experiment it is connected mainly with progress in the techniques of semiconductor detectors (Ge(Li) and Si(Li)). The high resolution of semiconductor detectors (better than 1 keV), makes it possible to separate experimentally many transitions between different shells of the atom, and this, together with their high efficiency (up to 10%), makes it possible to use these detectors where it is impossible to employ crystal spectrometers, which have a higher resolution but a much lower transmission ($\ll 1\%$).

In the present study we used a semiconductor Si(Li) spectrometer to determine the fluorescence yield ω_2 for the L_2 subshell of an atom with Z = 96. Insofar as we know, the fluorescence yields have been measured so far only for atoms with $Z \leq 94$. At the same time, measurement of the probability of the radiative transitions to the L sublevels of the heaviest atoms is of great theoretical interest, particularly in connection with the break (drop) in ω_2 predicted by Listengarten[7] for $Z \geq 91$, owing to the possible (energy-allowed) Koster-Kronig transitions of the type $L_2 - L_3M_{4,5}$.

We measured the number and the energy distribution of the quanta of the characteristic radiation of ^{248}Cm, which accompanies the α decay of ^{252}Cf. The measurements were made by the αX-coincidence method relative to the known L-emission yields in α decay of ^{241}Am[8]. To this end, a ^{252}Cf + ^{241}Am source, obtained from a mixture of these isotopes by sputtering in vacuum onto an aluminum substrate (of approximate thickness 6 mg/cm^2) was placed over a diffusion-drift Si(Li) detector (with the substrate facing the detector). Another semiconductor detector, Si(Au), was placed over the source and used to register the α particles. A differential discriminator was used to "cut out" successively from the source α-particle spectrum the lines of ^{241}Am or ^{252}Cf (approximate distance between lines 600 keV, experimental half-width 80 keV), and coincidence with these lines was used to plot the spectrum of the L radiation of the corresponding daughter elements. The Si(Li) x-ray spectrometer[9] had a resolution of 700 eV under the conditions of our investigation. The thickness of the dead layer and the depth of the sensitive band of the detector (which determine its efficiency) were obtained by comparing the measured relative intensities of the lines from ^{57}Co and ^{241}Am sources with the known published data[8,9]. They turned out to be 7 and 2450 μ, respectively.

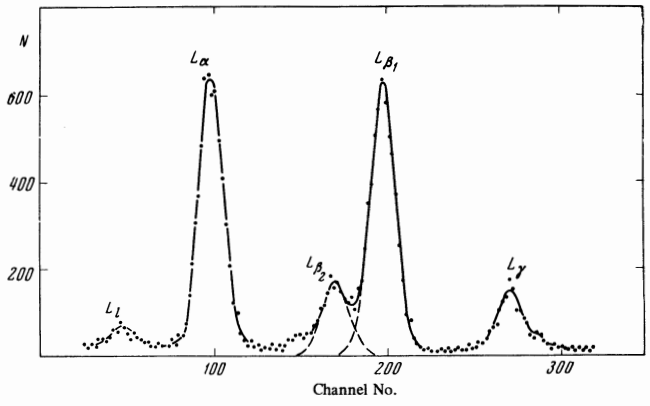
The so-measured energy distribution of the characteristic L radiation (curium) accompanying the α decay of ^{252}Cf is shown in the figure (without corrections for efficiency). Taking into account the absorption of the radiation in the substrate of the source (which amounted to less than 10% for the softest (L_l) series) and the efficiency of the detector (more than 75% for the hardest (L_γ) series), we determined, by integrating the spectrum, the number $N_{\alpha L}$ of the curium x-ray quanta corresponding to the registered number of α particles N_α. We also determined the same quantity $N^0_{\alpha L}$, corresponding to N^0_α, for ^{241}Am in the same manner. This made it possible to determine the yield F_L of the L radiation per α decay of ^{252}Cf from the relation

$$F_L = F_L^0 \frac{N_{\alpha L}/N_\alpha}{N_{\alpha L}^0/N_\alpha^0}$$

where F^0_L is the yield per α decay of ^{241}Am.

Under the condition that $F^0_L = 0.376 \pm 0.007$[8], taking into account the errors in F^0_L, the statistical errors, the errors in the calibration of the spectrometer, and the corrections for the random-coincidence background (7%), we obtained $F_L = 0.063 \pm 0.003$.

The characteristic L radiation observed in the α decay of ^{252}Cf is the result of internal conversions of tran-



Spectrum of L series of characteristic radiation of ^{248}Cm in the α decay of ^{252}Cf. The dashed lines show the results of the graphic separation of the lines L_{β_1} and L_{β_2}.

sitions from the first excited level of ^{248}Cm, with energy 43.1 keV, to the ground-state level (the contribution of transitions from higher levels can be neglected within the limits of the experimental accuracy). It has been well established that this is an E2 transition, just as for all even-even nuclei. In the absence of experimental data, knowledge of the energy and type of the transition makes it possible to calculate the fraction of the conversion coefficient α_L on the L shell and the total α conversion coefficient. From the tables of[11,12] we found that $\alpha_L/(\alpha+1) = 0.710$ ($\alpha_L/(\alpha+1) \approx \alpha_L/\alpha$, since $\alpha \sim 1000$); the error in this quantity is apparently approximately 5%. The number of vacancies produced per α decay in the curium L-shell is

$$n_L = W_{43}\alpha_L / (\alpha + 1),$$

where W_{43} is the relative probability of the α transition to the level with energy 43 keV. Since $W_{43} = 0.158$[13] (with an uncertainty on the order of 2%), it follows that $n_L = 0.112 \pm 0.006$. The number of x-ray quanta per vacancy, i.e., the average fluorescence yield for the L shell, is $\bar{\omega}_L = F_L/n_L$, and consequently $\bar{\omega}_L = 0.56 \pm 0.04$.

The average fluorescence yield depends on the distribution of the primary vacancies among the subshells, and therefore depends on the method of vacancy formation—the method of excitation of the fluorescence. The quantities of physical interest are not the average yields, but the fluorescence yields for the individual subshells ω_i, being the probabilities of vacancy filling in these subshells by radiative transitions. It is precisely for their determination that the high energy resolution of the L-radiation spectrometer is necessary. As shown in[14], in the case of pure E2 transitions, when the L_1-subshell ionization can be neglected, the fluorescence yield ω_2 for the L_2 subshell can be determined from the average value by experimentally separating the contributions made to the total L radiation by the sublevels L_2 and L_3 (F_2 and F_3) and finding their relative shares of vacancies (n_2 and n_3):

$$\omega_2 = \bar{\omega}_L(1 + n_3/n_2) / (1 + F_3/F_2).$$

The ratio n_3/n_2 of the primary vacancies was calculated from the corresponding conversion coefficients and found to equal 0.83, with an expected accuracy better than 2–3%[15].

The value of F_3/F_2 should be determined from experiment. The intensity of the x-ray lines connected with transitions to different subshells was previously measured with crystal spectrometers, and this called for prolonged exposures (for example, 112 days for plutonium[14]). The spectrum of the ^{248}Cm L radiation, shown in the figure, was obtained within 5 hours. More than 98% of the spectrum comes from transitions to the sublevels L_2 and L_3 (the more intense lines are marked in the figure). The boundary between the groups of L_2 and L_3 lines passes over the L_β series (the series L_l and L_α pertain entirely to L_3, while L_γ pertains entirely to L_2). The separation of L_{β_2} ($L_3 - N_5$) from L_{β_1} ($L_2 - M_4$), which was attained with our spectrometer, makes it possible to separate reliably the contributions of the two subshells and to find that $F_3/F_2 = 1.16 \pm 0.03$. Substituting all the obtained quantities in the expression for ω_2, we find that the fluorescence yield for the L_2 subshell is $\omega_2 = 0.48 \pm 0.04$.

Element	Z	Fluorescence yield for L_2 subshell
Th	90	0,56
U	92	0,59
Np	93	0,79±0,19
Pu	94	0,41±0,02
Cm	96	0,48±0,04

The table lists all the presently known values of ω_2 for nuclei with $Z \geq 90$. All the results for Z = 90–94 were taken from the review of Fink et al.[16], where the reference to the original papers can be found. A comparison of the data in the table apparently favors a certain decrease of the rate of radiative transitions to the L_2 subshell of the heaviest atoms.

[1] I. H. Scofield, Phys. Rev. **179,** 9 (1969).
[2] H. R. Rosner and C. P. Bhalla, Z. Phys. **231,** 347 (1970).
[3] E. I. McCuire, Phys. Rev. **185,** 1 (1969); Phys. Rev. A **2,** 273 (1970); Phys. Rev. A **3,** 587 (1971).
[4] G. C. Nelson and B. G. Sannders, Phys. Rev. **188,** 108 (1969).
[5] I. S. Hansen, H. U. Freund, and R. W. Fink, Nucl. Phys. A. **153,** 465 (1970).
[6] A. G. de Pinho, Phys. Rev. A **3,** 905 (1971).
[7] M. A. Listengarten, Izv. Akad. Nauk SSSR Ser. Fiz. **24,** 1041 (1960).
[8] L. B. Magnusson, Phys. Rev. **107,** 161 (1957).
[9] S. M. Solov'ev, L. I. Tyvin, and V. P. Éismont, Pribory i Tekhn. Éksperim. (in press).
[10] W. Rabinson and K. P. Gopinathan, Phys. Rev. **170,** 969 (1969).
[11] R. S. Hager and E. C. Seltzer, Nucl. Data Tables, A **4,** 1 (1968).
[12] O. Dragoun, H. C. Pauli, and F. Schmutzler, Nucl. Data Tables, A **6,** 235 (1969).
[13] S. A. Baranov, V. M. Shatinskiĭ, and V. M. Kulakov, Yad. Fiz. **11,** 701 (1970) [Sov. J. Nucl. Phys. **11,** 393 (1970)].
[14] L. Salgueiro, J. G. Ferreira, J. J. H. Park, and M. A. S. Ross, Proc. Phys. Soc. Lond. **77,** 657 (1961).
[15] I. H. Hamilton, Phys. Lett. **20,** 32 (1966).
[16] R. W. Fink, R. C. Jopson, Haus Mark, and C. D. Swift, Rev. Mod. Phys. **38,** 513 (1966).

Translated by J. G. Adashko
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