K and L X-Ray Intensities in Cesium-131 Decay*

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A measurement of the relative intensities of the K and L x-rays following electron-capture in Cs^{131} is reported. The result may be expressed $[L_I/K+L_{II,III}/K+n]\bar{\omega}_L=0.106\pm0.008$, where L_i/K is the ratio of L_i-shell capture to K-shell capture, n is the probability of an L-ionization following a K-vacancy, and $\bar{\omega}_L$ is the mean L-fluorescence yield of xenon. Upon combining this with recent theoretical results on electron-capture, the mean value of the L-fluorescence yield of xenon is found to be $0.10_3\pm0.01$. This value is suggested as a normalization point for the relative photographic values of $\bar{\omega}_L$ in the region $Z=54$.

In order to explain the observed anomalies in inner bremsstrahlung spectra of electron-capture on the basis of p-electron capture, one must conclude that L_{II}-capture is considerably more radiative than L_I-capture, and, furthermore, that the probability of radiative capture increases more strongly with Z for L_{II}-capture than for L_I-capture.

Traces of 6.2-day Cs^{132} and 13.6-day Cs^{136} were found in the first cesium fraction from pile-irradiated barium nitrate, presumably induced by (n,p) reactions.

INTRODUCTION

MARSHAK,[1] Rose and Jackson,[2] and Brysk and Rose[3] have computed theoretical values of the L_I/K-capture ratio as a function of atomic number and disintegration energy. From the curves of Brysk and Rose, the value of this ratio is computed to be 0.14 for Cs^{131}.

Saraf[4] has found an anomaly in the shape of the inner bremsstrahlung spectrum of Cs^{131} (which we have confirmed independently[5]); similar anomalies have been reported[6–8] in other electron capturers. Glauber and Martin[9] have suggested that the observed shape can be explained by taking into account the capture of electrons of nonzero angular momentum. This indicates that capture of L_{II} and L_{III} electrons might make a significant contribution to the inner bremsstrahlung.

The electron-capture of Cs^{131} is an allowed transition since $\log ft=5.3$ and the shell model predicts a $d_{5/2}\rightarrow d_{3/2}$ ($\Delta I=1$, no) transition.[4] In an attempt to investigate the relative contribution of p-electron capture, it seemed of interest to measure the ratio of the intensities of K and L x-rays following electron capture in Cs^{131}.

EXPERIMENTAL METHOD

A source was prepared by chemical separation of cesium from 44 grams of barium nitrate irradiated for 28 days in the Oak Ridge reactor. After precipitation of $BaCl_2\cdot H_2O$ from evaporation with (5:1) HCl-diethyl ether mixture, the supernatant cesium fraction was scavenged extensively with barium chloride precipitations, followed by lanthanum hydroxide, ferric hydroxide, and silver chloride scavengings. Separation from other alkali metals was accomplished by precipitation as the silicotungstate. Scintillation spectrometer studies of the final cesium fraction detected no trace of the intense gamma rays of Ba^{131}, indicating a decontamination factor from barium of better than 10^6. Traces of 6.2-day Cs^{132} and 13.6-day Cs^{136} were identified by scintillation spectrometry; these isotopes presumably are formed by (n,p) reactions in barium. Saraf[4] also observed Cs^{132} produced in this way. The traces of these isotopes have negligible effect on measurements reported here.

The L and K x-ray intensities were measured using an x-ray proportional counter filled with 2.1 atmos argon—0.2 atmos methane mixture. The counter was constructed of brass with an aluminum liner to eliminate fluorescent radiation from the brass. The side window was a 164 mg/cm² disk of Brush beryllium. The x-ray path length in the counter was about 10 cm. A potential of 3850 volts was applied to the 0.004-inch diameter stainless steel wire, and the amplified output was fed through a single-channel pulse analyzer into a recording ratemeter or scaler. A typical spectrum is shown in Fig. 1. The observed ratio of x-ray intensities, (N_L/N_K), was obtained by graphical integration using a planimeter. This ratio is $0.33_4\pm0.01$, representing an average of many determinations. The same value was also determined by the method of integral-bias counting, giving an independent check on the result. The intensity ratio may be expressed by Eq. (1)

$$\left(\frac{N_L}{N_K}\right)=\left[\frac{L_I+L_{II,III}+nK}{K}\right]\times\left(\frac{\bar{\omega}_L}{\omega_K}\right)\left(\frac{T_L}{T_K}\right)_W\left(\frac{T_L}{T_K}\right)_A\left(\frac{\xi_L}{\xi_K}\right)\left(\frac{S_L}{S_K}\right), \quad (1)$$

where K is the probability that a disintegration will

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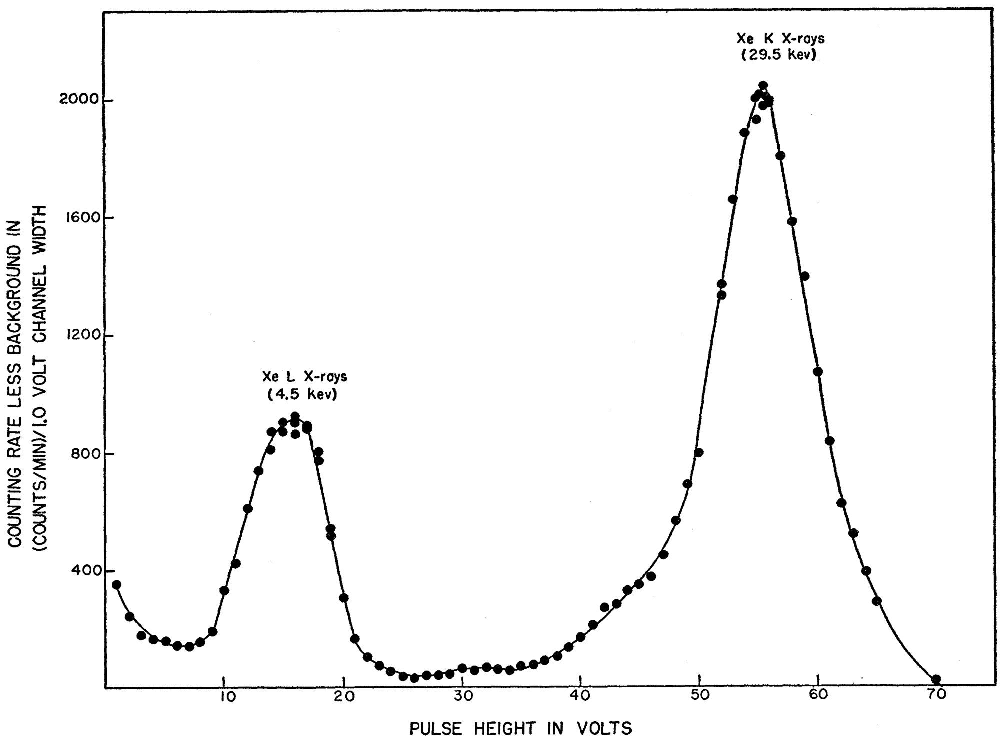

FIG. 1. Pulse-height spectrum of 9.8-day Cs^{131} in x-ray proportional counter filled with 2.1 atmos argon and 0.2 atmos methane. The points are those determined using a scaler and are corrected for background. The ratio of areas, (N_L/N_K), is $0.33_4\pm0.01$ by graphical integration. Integral-bias counting agrees exactly with this ratio within the above error limits.

occur by capture of an electron from the K-shell; $L_{\rm I}$, $L_{\rm II}$, and $L_{\rm III}$ are the probabilities of capture from the respective L-shells; n is the probability that a K-shell vacancy is filled by an L-electron; ω_K is the K x-ray fluorescence yield of xenon, taken as 0.87 ± 0.03 from the curve of Broyles, Thomas, and Haynes[10]; $\bar{\omega}_L$ is the mean L-fluorescence yield of xenon, which is discussed below; $(T_L/T_K)_W$ is the relative window transmission of xenon L and K x-rays, evaluated as 0.279 ± 0.006; $(T_L/T_K)_A$ is the relative air transmission which turns out to be very close to unity; (ξ_L/ξ_K) is the ratio of the gas counting efficiencies of xenon L and K x-rays in argon, which is 9.9 ± 0.5, computed from mass absorption coefficients from Allen[11]; and (S_L/S_K) is the correction for the self-absorption and self-scattering of x-rays in the source, taken as unity in the first approximation.

TABLE I. Xe L-shell ionization probabilities following K-shell ionizations.

Shell	Radiative transition	Auger transition	Total
$L_{\rm I}$	0.00	0.07	0.07
$L_{\rm II}$	0.23_5	0.04_5	0.28
$L_{\rm III}$	0.47	0.06_5	0.53
$L_{\rm total}$	0.70	0.18	$0.88=n$

[10] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).
[11] S. J. M. Allen, tables in *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, 1952), thirty-fourth edition.

Equation (1) may be rearranged and evaluated to give the quantity

$$[L_{\rm I}/K+L_{\rm II,\,III}/K+n]\bar{\omega}_L=0.106\pm0.008, \quad (2)$$

which is our experimental result.

INTERPRETATION AND DISCUSSION

1. Ratio of $L_{\rm I}$ to K Capture

Brysk and Rose[3] have shown that for an allowed transition

$$L_{\rm I}/K=g_{L{\rm I}}^2q_{L{\rm I}}^2/g_K^2q_K^2, \quad (3)$$

where $g_{L{\rm I}}$, g_K are screened relativistic Coulomb wave functions evaluated at the nuclear surface, and $q_{L{\rm I}}$, q_K are the neutrino energies associated with $L_{\rm I}$ and K capture, respectively. For Cs^{131}, q_K is 320 ± 10 kev from inner bremsstrahlung measurements.[4,5] From the relativistic wave-function ratios of Rose and Jackson[2] and from the graphs of Brysk and Rose, the value of $L_{\rm I}/K$ is found to be 0.14.

2. Ratio of $L_{\rm II}$ and $L_{\rm III}$ to K Capture

Brysk and Rose have shown that $L_{\rm III}$ capture always is negligible for allowed and first-forbidden $(\Delta I=0, 1)$

transitions because the neutrino is forced into a higher angular momentum state. From their curves one finds that $L_{II}/L_I = 0.027$ in the case of Cs^{131} and thus $L_{II}/K = 0.004$.

3. L-Ionization Following K-Ionization

The probability that a K-shell vacancy is filled by an L-electron can be calculated from x-ray and Auger line intensities. The values of n have been interpolated from Burhop[12] and Massey and Burhop,[13] and are given in Table I. Since the K-fluorescence yield of xenon is large, the greatest contributions to n arise from radiative transitions, the intensities of which are well known.[13] The contribution of Auger transitions has been taken from the relative intensities of Auger lines at higher Z.[14] Coster-Krönig transitions, in which L-vacancies are shifted from one subshell to another, do not occur for $50<Z<75$.

4. L-Fluorescence Yield of Xenon

Experimental determinations of $\bar{\omega}_L$ have been made by Auger and by Bower, using cloud-chamber techniques, and by Lay, using a photographic method.[12] Their values are summarized in Table II, and are too large to be consistent with the work reported here. Using the values for L_I/K, L_{II}/K, L_{III}/K, and n deduced in the aforementioned, we find that $\bar{\omega}_L = 0.10_3 \pm 0.01$ for xenon.

CONCLUSION

Combining the theoretical values of Brysk and Rose[3] for L-capture with our experimental x-ray intensity ratio, we have determined the mean L-fluorescence yield of xenon:

$$\bar{\omega}_L = 0.10_3 \pm 0.01.$$

Lay's values,[15] obtained by making photographic comparisons between the intensities of incident and fluorescent x-ray beams for $40<Z<92$, are considered to be internally consistent; our absolute value might serve as a normalization point, at least in the region of $Z=54$.

If the observed inner bremsstrahlung anomaly[4–8] is attributed to radiative capture of p-electrons,[9] then a comparison of the intensity of the anomalous radiation with the theoretical probability of L_{II}-capture leads to to the conclusion that L_{II}-capture must be considerably more radiative than L_I-capture. Comparing the spectrum[7] of Fe^{55} with that[4,5] of Cs^{131}, one further concludes that the probability of radiative capture increases more strongly with Z for L_{II}-capture than for L_I-capture.

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APPENDIX

Calculation of $\bar{\omega}_L$ for Xenon

Following the method of Kinsey,[16] the fluorescence yield of the L levels of xenon have been computed separately on the basis of radiation widths obtained by interpolation from the work of Massey and Burhop,[13] and the existing information on the widths of L-levels. The method depends on the fact that (if Coster-Krönig transitions are absent)

$$\omega = \Gamma_{rad}/\Gamma_{total}, \tag{4}$$

where ω is the fluorescence yield of a given level, Γ_{rad} is the level width for radiation, and Γ_{total} is the total level width.

The total widths can be measured by studying the shapes of the absorption edges of the L levels. Green's[17] values for barium and iodine, and Sandström's[18] values for tellurium and antimony are shown in Table III, along with our assumed values for xenon. Sandström shows that widths vary only slowly with Z. The radiation widths of the L levels, obtained by interpolating between values of Massey and Burhop for anti-

TABLE II. L-fluorescence yields of xenon.

$\bar{\omega}_L$	Investigator	Remarks
0.25	Auger[a]	Using Xe-filled cloud chamber
0.21	Bower[b]	Using Xe-filled cloud chamber
(0.13)	Lay[c]	Photographic comparison of incident to fluorescent beams (as interpolated by present authors).
0.03	Present authors	Calculated according to the method of Kinsey[d] (see Appendix).
$0.10_3 \pm 0.01$	Present authors	From present experiment.

[a] P. Auger, J. phys. radium 6, 205 (1925).
[b] J. C. Bower, Proc. Roy. Soc. (London) A157, 662 (1936).
[c] H. Lay, Z. Physik 91, 533 (1934).
[d] See reference 16.

TABLE III. L-level widths in the region of xenon and partial L-fluorescence yields.

Level	Γ_{total}(ev) Sb[a]	Te[a]	I[b]	Ba[b]	Xe[c]	Γ_{rad}(ev)[d]	ω
L_I	5.6	4.9			5.2	0.09	0.017
L_{II}	4.7	4.6			4.7	0.12	0.025
L_{III}	4.1	4.7	2.6 ± 0.1	≤ 4.06	4.2 2.8	0.10	0.023 0.036
						Weighted mean	$\bar{\omega}_L = 0.03 \pm 0.01$

[a] See reference 18.
[b] See reference 17.
[c] Assumed by present authors.
[d] Interpolated from reference 13.

[12] E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, 1954), p. 50 ff.
[13] H. W. S. Massey and E. H. S. Burhop, Proc. Cambridge Phil. Soc. 32, 461 (1936).
[14] See reference 12, page 61.
[15] H. Lay, Z. Physik 91, 533 (1934).
[16] B. B. Kinsey, Can. J. Research A26, 404 (1948).
[17] E. H. Green, Phys. Rev. 55, 1072 (1939).
[18] A. E. Sandström, Phil. Mag. 22, 497 (1936).

mony and gold, are also shown in Table III. Values of the separate L-fluorescence yields are shown in the last column of Table III. From these values and the values of n in the last column of Table I, we compute a weighted mean fluorescence yield for xenon, $\bar{\omega}_L = 0.03$.

Fluorescence yields calculated in this manner by Kinsey for high Z are in general somewhat lower than values found experimentally. For xenon the value is considerably lower and is inconsistent with the present experiment.

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Photoneutron Cross Sections in Mg^{24}, Mg^{25}, Zr^{90}, Zr^{91}†

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The variation of $\sigma(\gamma,n)$ with E_γ from threshold to 24 Mev has been measured by detecting neutrons produced from betatron bremsstrahlung irradiation of separated isotope samples in oxide form. Despite the large threshold differences (9.3 Mev for the Mg isotopes and 5.0 Mev for the Zr isotopes) the peaks of the giant dipole resonances are very close for each pair of adjacent isotopes: Mg^{24}, 19.5 Mev; Mg^{25}, 20.3 Mev; Zr^{90}, 16.4 Mev; Zr^{91}, 16.3 Mev. The half-widths of the Zr resonances are unusually small, 4.1 and 5.4 Mev, compared to 8 Mev for nearby nuclei observed previously. This is apparently a "magic number" effect.

INTRODUCTION

PREVIOUS measurements in this laboratory[1] have indicated that the giant dipole resonances in (γ,n) processes occur at a photon energy which depends only on the mass number of the nuclide. The energy (E_m) at which the maximum cross section occurs varies slowly from about 13.5 Mev for $A=200$ to about 22.5 Mev for $A\leq 20$. Measurements elsewhere[2] have generally been in accord with this observation, but deviations from the smooth variation have been reported. It has been suggested[3] that the apparent smooth variation of E_m with A is accidental and that E_m is instead closely related to the (γ,n) threshold energy, E_{th}, so that $E_m - E_{th}$ is approximately constant. Since E_{th} does in general decrease slowly with increasing A, this is not in absolute contradiction to our observations.

A second interesting feature of the giant resonances which was observed here previously[1] is that the width of the resonance varies slowly from about 7 Mev for $A=238$ to 10 Mev for $A=31$. (Below $A=31$ the widths for the 5 elements we have measured so far are much smaller, 3 to 7 Mev.) However, the nuclides with 28, 52, 82, and 126 neutrons have widths from 2 to 4 Mev less than the smooth variation noted for other nuclides. Except for $N=52$ these are "magic" neutron number nuclides, and $N=52$ is only two units away from the magic number 50.

In making a careful study of the variation of E_m with A or with E_{th}, the neutron detection method is to be preferred over the method of residual activity because the apparatus is identical for all target materials, and problems of half-lives, decay schemes, and decay energies differing for the various nuclides used do not enter. (It is also necessary to use only data from a single laboratory in making a critical evaluation, because of the known difficulties in comparing photonuclear cross sections measured in different laboratories.[4]) There is however the limitation that one is restricted to measurements on isotopes of not much less than 100 percent abundance. It is thus possible that observed variations of E_m with A might be of a special nature, related to the properties of the relatively small number of nuclides which can be studied in their naturally occurring abundances. This limitation can be overcome by using separated isotopes, which are now rather extensively available from the Oak Ridge National Laboratory.

A critical test of the dependence of E_m on E_{th} or on A can be made by measuring the (γ,n) cross section *vs* energy curve for two adjacent isotopes which have very different E_{th} values. On the hypothesis that the mass number is the determining factor, E_m will be the same for the two isotopes. On the hypothesis that E_{th} is the determining factor, the difference in the E_m values for the two isotopes should be comparable to the difference in the E_{th} values.

One of the most extreme cases available for examination is the isotopic pair Mg^{24}-Mg^{25}. The natural abun-

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[3] L. Katz and A. S. Penfold, Phys. Rev. **81**, 815 (1951); Katz, Baker, Haslam, and Douglas, Phys. Rev. **82**, 271 (1951).
[4] J. Goldemberg and L. Katz, Can. J. Phys. **32**, 49 (1954), discuss the degree of agreement among various laboratories engaged in this work.