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L-shell internal ionization in the beta decay of ^{203}Hg†

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L-shell internal ionization has been observed in the β decay of ^{203}Hg. A spectrum of Tl L x rays was obtained from a 0.12-μC source of ^{203}Hg in coincidence with 279-keV x rays from ^{203}Tl. The coincidence requirement served to eliminate internal-conversion x rays from the spectrum. The total L-shell internal ionization probability P_L obtained after normalization to the measured singles x-ray intensity, was found to be $(12.9 \pm 0.5) \times 10^{-4}$. Estimates for the individual L-subshell internal ionization probabilities resulted in values of $(0.0 \pm 6.5) \times 10^{-4}$, $(4.8 \pm 2.3) \times 10^{-4}$, and $(8.1 \pm 3.5) \times 10^{-4}$ for $P_{L_{\rm I}}$, $P_{L_{\rm II}}$, and $P_{L_{\rm III}}$, respectively. The measured value of P_L is over 4 standard deviations higher, and that of $P_{L_{\rm III}}$ over 1 standard deviation higher, than theoretical predictions.

[RADIOACTIVITY ^{203}Hg; measured γ-x-ray coinc; deduced internal ionization probabilities P_L, $P_{L_{\rm I}}$, $P_{L_{\rm II}}$, $P_{L_{\rm III}}$.]

INTRODUCTION

In recent years, the interaction between atomic and nuclear processes has attracted considerable attention. One particular quantity of interest is the probability that changes will occur in the electronic configuration of an atom during the β decay of its nucleus.

Experimentally, the easiest of these configuration changes to measure is K-shell ionization, which results in the emission of K x rays characteristic of the daughter element. Measurements of the probability P_K per beta decay for this process, called K-shell "autoionization" or "internal ionization" have been made on over a dozen nuclides throughout the Periodic Table. A recent compilation of these results is presented in Ref. 1. Most of the nuclides selected for study decay through ground-state-to-ground-state transitions, as transitions to excited daughter states give rise to internal conversion x rays in the daughter atom which are indistinguishable from x rays due to internal ionization. It has been shown,[2] however, that internal ionization x rays can be measured in some nuclei which decay to excited daughter states by means of a γ-x-ray coincidence technique.

Some theoretical calculations[3,4] have been extended to include the probability P_L of L-shell internal ionization and even the partial probabilities of internal ionization in the three L subshells. Total L-shell internal ionization probabilities have been measured for five nuclides, ^{90}Y, ^{143}Pr, ^{147}Pm, ^{204}Tl, and ^{210}Bi. A review of recent work is found in Ref. 4.

In the case of ^{204}Tl, Howard, Seykora, and Waltner[5] have attempted to deduce the separate partial probabilities of internal ionization in the three L subshells from the experimental data. The authors encountered difficulties in data reduction because of ambiguities in the L subshell fluorescence yields and Coster-Kronig transition rates. Taking these difficulties into account, however, the deduced distribution of internal ionization probability among the three L shells is still in considerable disagreement with available theoretical calculations.

We present here measurements of both the total probability of L-shell internal ionization in the β decay of ^{203}Hg and the partial internal ionization probabilities in the three L subshells. A γ-x-ray coincidence technique was employed (see Ref. 2). The L x-ray spectrum of Tl was measured in coincidence with the 279-keV γ ray to the ground state of ^{203}Tl. The decay scheme is shown in Fig. 1. A value of P_L is obtained using the singles intensity of Tl L x rays for normalization. From the

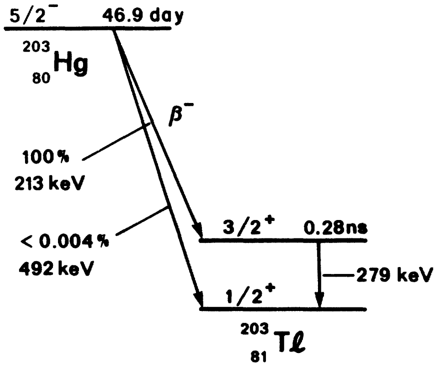

FIG. 1. Decay scheme of ^{203}Hg.

separate intensities of the $L\alpha$, $L\beta$, and $L\gamma$ peaks, the partial L-shell internal ionization probabilities $P_{L_{\rm I}}$, $P_{L_{\rm II}}$, and $P_{L_{\rm III}}$ are deduced, and the results are compared with the theoretical calculations of Carlson *et al.*[3] and Law and Campbell.[4]

EXPERIMENTAL PROCEDURE

A ^{203}Hg source of about 0.12 μC strength was prepared by evaporating a solution of mercuric nitrate in HNO_3 onto a film of Mylar 0.0013 cm in thickness. The source was placed between a Ge(Li) x-ray detector and a NaI(Tl) detector, using polyethylene disk mounts. The NaI(Tl) detector system consisted of a 7.6× 7.6 cm NaI(Tl) crystal coupled to an RCA 8575 phototube. The Ge(Li) detector was 5 mm thick and 16 mm in diameter, and was connected to a low noise charge-sensitive preamplifier. The source was situated 1.3 cm from the front of the Na(Tl) detector and 1.8 cm from the front of the Ge(Li) detector. Because of the appreciable flux of iodine K x rays excited in the NaI(Tl) detector, an aluminum plate 3.2 mm in thickness was interposed between the NaI(Tl) detector and the source, in order to cut down the intensity of these x rays seen by the Ge(Li) detector. In addition, to shield the Ge(Li) detector from the beta particles, a plate of beryllium 1 mm in thickness was placed between the source and the detector.

A slow-fast coincidence system was used for the experiment. Slow pulses from the NaI(Tl) detector were gated around the 279 keV γ-ray peak in a window 133 keV wide. Pulses from the Ge(Li) detector were gated from about 5 to 35 keV. A slow coincidence was formed between these two sets of gated pulses.

In the fast-coincidence system, pulses from the output of the Ge(Li) preamplifier, and the fast signals from the RCA 8575 phototube were each passed through a timing filter amplifier and a fast discriminator, the outputs of which were then fed into a time-to-amplitude converter (TAC). Output pulses from the TAC were gated with the pulses from the slow-coincidence system. The timing peak exhibited a full width at half maximum of about 40 nsec. The gated TAC pulses were passed through two pulse height analysers (PHAs), each with a window of 110 nsec. One was centered on the timing peak while the other was centered 460 nsec off the peak. The output of these PHAs gated two multiplexed inputs to a Northern Scientific 8192-channel analyzer. One input measured the x-ray spectrum corresponding to true-plus-random coincidences, while simultaneously the other was measuring the spectrum corresponding to random coincidences only. The total true-to-random ratio was about 25 : 1.

The coincidence data were collected for a total time of 169 h, in four separate runs. The singles x-ray spectra were collected, for 1024 sec each time, before and after each coincidence run. These singles served both as a check for shift in gain in the x-ray branch of the coincidence and as normalization for coincidence data. No shift in gain of more than one-half channel (44 eV) was detected during the experiment. In addition, the TAC spectrum and the singles spectrum from the NaI (Tl) detector were monitored at frequent intervals during the experiment to ensure that the timing and γ-ray energy gates remained centered on their respective peaks.

DATA ANALYSIS

Figure 2 shows the Ge(Li) coincidence spectrum after subtraction of randoms. This spectrum was subsequently corrected for several effects. The efficiency of the Ge(Li) detector, for example, undergoes an abrupt drop of about 15% as the energy increases above 11 keV because of the Ge K x-ray escape peak, and thereafter rises with energy at the rate of about 1.6% per keV in the region of interest. The data were corrected for this effect using the efficiency measurements of Forcinal,[6] in order to make the counting rate uniformly proportional to x-ray intensity for all channels in the region of interest.

The continuous background in the coincidence spectrum is assumed to be due principally to bremsstrahlung (both internal and external) associated with β-decay events. This background appears to be linear in an interval of 11.3 keV around the Tl L x rays. The background was estimated by taking averages of the measured intensities

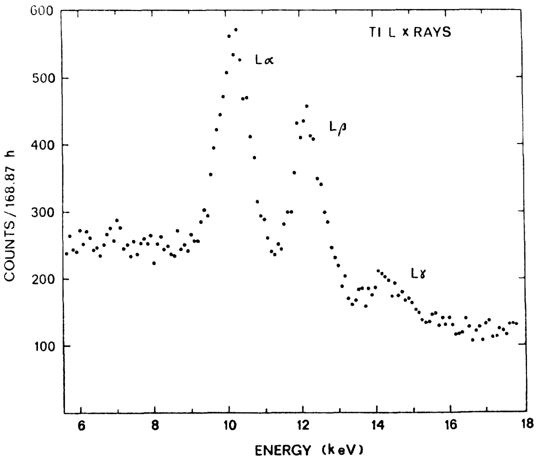

FIG. 2. L x-ray spectrum from the ^{203}Hg source in coincidence with 279 keV x rays. Random coincidences have been subtracted from the data.

between 6.3 and 8.0 keV and between 15.4 and 17.6 keV and connecting these averages with a straight line. The spectrum was corrected for the transmission of the x rays through the beryllium plate between the source and the Ge(Li) detector.

The singles L x-ray spectrum was similarly corrected for the variation of Ge(Li) efficiency with energy, for background subtraction and for transmission. In addition, corrections were made for the finite source lifetime to insure that the effective average source intensity for the singles measurements was the same as for the coincidence measurements.

The requirement that the spectrum of Fig. 2 be taken in coincidence with γ rays to the ground state of ^{203}Tl eliminates the contribution to the spectrum of internal conversion x rays from Tl. The coincidence spectrum thus consists of x rays resulting from L vacancies caused either by L-shell internal ionization or by the transfer of L electrons to K vacancies created by K-shell internal ionization. The K-shell internal ionization probability P_K in the β decay of ^{203}Hg has recently been measured,[2] and is about 10^{-2} times smaller than the present result for P_L, so that within the experimental errors, the creation of L-shell vacancies in coincidence with 279 keV γ rays can be attributed exclusively to the L-shell internal ionization process. The coincidence counting rate R_c integrated over all L x-ray peaks is therefore given by the expression

$$R_c = P_L r_\beta \bar{\omega}_L D_x \left(\frac{1}{1+\alpha}\right) D_\gamma, \tag{1}$$

where P_L is the probability per β decay of L-shell internal ionization, r_β is the decay rate of the source, $\bar{\omega}_L$ is the average L-shell fluorescence yield, D_x is the fraction of emitted L x rays detected by the Ge(Li) detector, D_γ is the fraction of emitted γ rays detected by the NaI(Tl) detector, and α is the total conversion coefficient for the 279 keV transition.

Normalization of R_c to the singles L x-ray counting rate has the advantage of eliminating several of these factors in obtaining P_L. Three processes contribute to the production of L-shell vacancies which result in singles L x rays:

(1) Internal conversion electrons are ejected from the L shell during the 279 keV transition;
(2) L electrons fill K vacancies resulting from K-shell internal conversion; and
(3) L-shell vacancies are produced by internal ionization as in the coincidence measurements.

The third effect can be estimated on the basis of the current measurement to be about 1% of the detected singles rate, and is therefore negligible compared to the first two effects. The singles x-ray counting rate is therefore given by the expression

$$R_x = r_\beta \bar{\omega}_L D_x \left(\frac{\alpha_L}{1+\alpha} + \frac{\alpha_K n_{KL}}{1+\alpha}\right), \tag{2}$$

where the two terms in parenthesis correspond to the first and second processes, respectively. Here α_L and α_K are the L- and K-shell conversion coefficients, and n_{KL} is the fraction of L vacancies created per K vacancy in the second process. From the above two expressions P_L may be found:

$$P_L = \frac{R_c}{R_x} \frac{(\alpha_L + \alpha_K n_{KL})}{D_\gamma}. \tag{3}$$

The values of α_L and α_K were 0.0475 and 0.1653, respectively, as taken from Hansen and Mouchel[7] and the value of n_{KL} used was 0.808 from Venugopala Rao, Chen, and Crasemann.[8] The final value of D_γ was 0.173, obtained from Heath[9] and corrected for transmission.

It should be noted that Eq. (3) is valid only if $\bar{\omega}_L$ is the same in the singles measurement as in the coincidence measurement. This is not obviously true, as the L-shell primary vacancy distributions in the two cases are not expected to be identical. However, it has been noted by Chen, Crasemann, and Kostroun[10] and by McGuire[11] that $\bar{\omega}_L$ is relatively insensitive to even drastic changes in the primary vacancy distribution. This fact was verified in the present case by calculating $\bar{\omega}_L$ assuming different primary vacancy distributions which corresponded to best estimates for the actual singles and coincidence distributions. Values of $\bar{\omega}_L$ so obtained differed from each other by less than 2%.

In order to estimate the partial L subshell vacancy distribution it was necessary to separate the L x-ray spectrum into three separate intensity groups: The lowest energy group, including the Ll and $L\alpha$ peaks, the intermediate group, including $L\eta$ and $L\beta$, and the highest energy group, including $L\gamma$. For convenience, these three groups will be referred to simply as $L\alpha$, $L\beta$, and $L\gamma$. Because of detector resolution, there was a small amount of overlap between the $L\alpha$ and $L\beta$ groups. This was corrected for by assuming the high energy wing of the $L\alpha$ peak to be Gaussian and subtracting this estimated contribution from the $L\beta$ group. The total amount of this contribution was less than 2% of the counts in the $L\beta$ group.

The relationship between $R_{L\alpha}$, the coincidence counting rate in the $L\alpha$ peak, and $I_{L\alpha}$, the total $L\alpha$ x ray intensity in coincidence with β decay events, is given by the expression

$$R_{L\alpha} = I_{L\alpha} D_x \left(\frac{1}{1+\alpha}\right) D_\gamma. \tag{4}$$

TABLE I. Subshell fluorescence yields, Coster-Kronig transition probabilities, and radiative branching ratios used in the present calculations.

	Value	Reference
ω_1	0.07 ± 0.02	14
ω_2	0.319 ± 0.010	14
ω_3	0.306 ± 0.010	14
f_{12}	0.14 ± 0.03	14
f_{13}	0.56 ± 0.05	14
f_{23}	0.113 ± 0.011	15
S_1	0.353 ± 0.053	12
S_2	0.238 ± 0.012	12
S_3	0.229 ± 0.012	14

Similar expressions relate $R_{L\beta}$ and $R_{L\gamma}$ to $I_{L\beta}$ and $I_{L\gamma}$, respectively. The intensity of L x rays X_1, X_2, and X_3 which result from the filling of final $L_{\rm I}$, $L_{\rm II}$, and $L_{\rm III}$ subshell vacancies, respectively, are related to the above total intensities through the equations

$$X_1 = (S_2S_3I_{L\alpha} - S_2I_{L\beta} + I_{L\gamma})(1+S_1)/(S_1-S_2), \tag{5}$$

$$X_2 = (-S_1S_3I_{L\alpha} + S_1I_{L\beta} - I_{L\gamma})(1+S_2)/(S_1-S_2), \tag{6}$$

$$X_3 = I_{L\alpha}(1+S_3), \tag{7}$$

where S_1, S_2, and S_3 are branching ratios associated with the filling of $L_{\rm I}$, $L_{\rm II}$, and $L_{\rm III}$ vacancies, respectively, and are defined in Ref. 12.

The rates of creation of primary vacancies N_1, N_2, and N_3 in the three subshells, respectively, are related to X_1, X_2, and X_3 by the expressions

$$N_1 = X_1/\omega_1, \tag{8}$$

$$N_2 = X_2/\omega_2 - (f_{12}/\omega_1)X_1, \tag{9}$$

$$N_3 = X_3/\omega_3 - (f_{23}/\omega_2)X_2 - (f_{13}/\omega_1)X_1, \tag{10}$$

where the ω_i's are the L-subshell fluorescence yields and the f_{ij}'s are the Coster-Kronig transition probabilities.

Finally, the probability per β decay of creation of a primary vacancy in the ith subshell P_{Li} is related to the primary vacancy creation rate in the ith subshell by the expression

$$P_{Li} = N_i/r_\beta. \tag{11}$$

These probabilities may be calculated in a straightforward manner from a knowledge of $R_{L\alpha}$, $R_{L\beta}$, and $R_{L\gamma}$. The measurement of R_x and the use of Eq. (2) eliminates the dependence of the calculation on D_x and r_β.

The calculation is, however, dependent on the values chosen for the S_i's, ω_i's, and f_{ij}'s. These quantities have been measured by various workers and have been compiled in a recent review article by Bambynek *et al.*[13] Many of the values obtained by different workers differ from each other by more than the quoted errors. In deciding which values to use, it was noted that the values deduced for several of these quantities were the result of calculations involving assumed values for others of the quantities, e.g., quoted values of f_{23} and S_2 depended upon the value chosen for S_3. In view of the interdependence of the values of these quantities, it was decided to use a consistent set of values measured by one group of workers. The set chosen was that obtained by Wood and coworkers[12,14,15] and listed in Table I. The dependence of the results on this choice was investigated by replacing the values of ω_2, ω_3, and f_{23} by those of Mohan.[13,16] In all cases, the resulting changes in the P_{Li} were small compared to the quoted errors in the measurements.

It should be noted that $\bar{\omega}_L$ does not cancel out in the calculation of the P_{Li} as it does in the calculation of P_L, and thus a value must be chosen for this quantity. Several independently measured values of $\bar{\omega}_L$ exist in the literature and are compiled in Ref. 13. In the present calculation it was decided not to use any of these measurements, but instead to calculate $\bar{\omega}_L$ from the set of values in Table I. The need for consistency between the values in Table I and $\bar{\omega}_L$ is forced by the use of the singles x rays to normalize the coincidence data. This procedure also results in a consistency between P_L and the sum of individual P_{Li}. In addition, the dependence of the calculated $\bar{\omega}_L$ on the ω_i and f_{ij} lessens the dependence of the P_{Li} on variations in these quantities.

RESULTS

The experimental results are presented in Table II. The errors include both contributions from the present data and quoted errors from the quantities listed in Table I. In all cases the principle contribution to the errors was from $I_{L\gamma}$. The very large error in $P_{L_{\rm I}}$ arises from the fact that the terms in the numerator of Eq. (5) almost exactly cancel each other.

TABLE II. Summary of experimental and theoretical results.

		Theoretical results × 10^4	
	Experimental results × 10^4	Carlson *et al.* (Ref. 3)	Law and Campbell (Ref. 4)
P_L	12.9 ± 0.5	10.59	10.5
$P_{L_{\rm I}}$	0.0 ± 6.5	4.47	3.8
$P_{L_{\rm II}}$	4.8 ± 2.3	2.74	3.1
$P_{L_{\rm III}}$	8.1 ± 3.5	3.38	3.6

The results of theoretical calculations by Carlson *et al.*,[3] and Law and Campbell[4] are also presented in Table II. The results of Carlson were obtained using a sudden approximation model for the process, and Hartree-Fock-Slater wave functions for the initial and final atomic states. The calculations of Law and Campbell involve a one-step model in which the indistinguishability of the two continuum electrons in the final state is taken into account explicitly. Dirac single-particle wave functions were employed in the calculations.

Comparison with the calculations indicates that the experimental values for $P_{L_{III}}$ and P_L are both somewhat higher then the theoretical predictions. Howard *et al.*,[5] have noticed similar discrepancies in the measurement of autoionization probabilities in the beta decay of ^{204}Tl. At present, there is no obvious explanation for these discrepancies.

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[1] J. Law and J. L. Campbell, Phys. Rev. C 12, 984 (1975).

[2] A. Bond, O. P. Gupta, and A. Zide, Phys. Rev. C 9, 1529 (1974).

[3] T. A. Carlson, C. W. Nestor, Jr., T. C. Tucker, and F. B. Malik, Phys. Rev. 169, 27 (1968).

[4] J. Law and J. L. Campbell, Nucl. Phys. A187, 525 (1972).

[5] J. M. Howard, E. J. Seykora, and A. W. Waltner, Phys. Rev. A 4, 1740 (1971).

[6] G. Forcinal, Nucl. Instrum. Methods 107, 125 (1973).

[7] H. H. Hansen and D. Mouchel, Z. Phys. 267, 371 (1974).

[8] P. Venugopala Rao, M. H. Chen, and B. Crasemann, Phys. Rev. A 5, 997 (1972).

[9] R. L. Heath, USAEC Report No. IDO-16880-1, 1964 (unpublished).

[10] M. H. Chen, B. Crasemann, and V. O. Kostroun, Phys. Rev. A 4, 1, (1971).

[11] E. J. McGuire, Phys. Rev. A 3, 587 (1971).

[12] P. Venugopala Rao, J. M. Palms, and R. E. Wood, Phys. Rev. A 3, 1568 (1971).

[13] W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, Hans Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, Rev. Mod. Phys. 44, 716 (1972).

[14] R. E. Wood, J. M. Palms, and P. Venugopala Rao, Phys. Rev. 187, 1497 (1969).

[15] V. R. Veluri, R. E. Wood, J. M. Palms, and P. Venugopala Rao, J. Phys. B 7, 1486 (1974).

[16] S. Mohan, Ph.D. thesis, Georgia Institute of Technology (1971) (unpublished).