

**$L/K$ -Capture Ratio of  $\text{Zn}^{65}$ †\***

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The  $L/K$ -capture ratio of  $\text{Zn}^{65}$  was determined to be  $0.119 \pm 0.007$  in a high-pressure, multiwire, gas proportional counter with radioactive zinc dialkyl as the gaseous source. This value is significantly larger than the theoretical value of 0.097 predicted by Brysk and Rose.

**INTRODUCTION**

THE most accurate ( $\pm 3$ –6%) experimental values for the ratio of the probability of  $L$ -electron capture to  $K$ -electron capture,  $P_L/P_K$ , have been determined at the lowest atomic numbers ( $17 < Z < 37$ ) by the high-pressure, multiwire, proportional counter technique where the error due to  $K$  x-ray escape—the escape peak is not resolvable from the  $L$  peak—is minimized by surrounding the center counter with a ring of counters connected in anticoincidence.<sup>1–5</sup> These values have been found to be larger than the theoretical values of  $P_L/P_K$  calculated over the atomic number range,  $3 < Z < 21$ , by Rose and Jackson<sup>6</sup> and over the range,  $13 < Z < 96$ , by Brysk and Rose.<sup>7</sup> When Odier and Daudel<sup>8</sup> considered the correlations between the positions of the atomic electrons in their theoretical calculation of  $P_L/P_K$ , they obtained a value about twice as large as Rose's for Be ( $Z=4$ ) and 22% larger, for Ar ( $Z=18$ ). Odier and Daudel's value of  $P_L/P_K$  for Ar, the heaviest element they considered, is in excellent agreement with the experimental results.<sup>4</sup> This positive discrepancy between the experimental values and the Brysk and Rose values is smaller in the first transition series, where it amounts to an average of 10% for the five nuclides thus far measured (manganese-54,<sup>5</sup> iron-55,<sup>3,5</sup> cobalt-58,<sup>5</sup> germanium-71,<sup>1</sup> and krypton-79<sup>1</sup>). Another nuclide which can be used in establishing the magnitude of the discrepancy in this region is  $\text{Zn}^{65}$ .

Townsend<sup>9</sup> calculated the  $L/(K+L)$ -capture ratio, i.e.,  $P_L/(P_K+P_L)$ , of  $\text{Zn}^{65}$  to be  $0.08 \pm 0.04$  from the experimental value of the apparent ratio extrapolated to zero pressure, where all the  $K$  x rays were assumed to escape his proportional counter. As previously noted,<sup>1</sup>

the ratio can not be determined very accurately in such a low-pressure experiment as it is critically dependent on the value chosen for the  $K$ -fluorescence yield,  $\omega_K$ .

Recently, Perrin<sup>10</sup> undertook to measure the  $(L+M+\dots)/K$ -capture ratio of  $\text{Zn}^{65}$  by employing x- $\gamma$  coincidence techniques using two NaI(Tl) scintillation crystals. However, the calculation involves a small difference between two large numbers. Therefore, the result is strongly dependent on the correction for air and window absorption of the x rays and on the value of  $\omega_K$ . Thus, by using a  $\omega_K$  of 0.45 for Cu,<sup>11</sup> Perrin found  $(P_L+P_M+\dots)/P_K$  to be  $0.22 \pm 0.06$ . Using the theoretical value of  $(P_M+P_N+\dots)/P_L$ ,<sup>1</sup>  $P_L/P_K$  is found to be  $0.19 \pm 0.05$ . However, if the most recent value<sup>12</sup> of 0.3929 for the  $\omega_K$  of Cu were used,  $P_L/P_K$  would be 0.06.

In view of the large limits of error in the previous studies it was decided to investigate  $\text{Zn}^{65}$  in a multiwire, proportional counter. This nuclide decays to the ground state of Cu<sup>65</sup> by electron capture, positron emission, and electron capture followed by a  $1.118 \pm 0.005$ -MeV  $\gamma$  ray.<sup>13</sup> From the end-point energy of the positron spectrum,  $0.324 \pm 0.002$  MeV,<sup>13</sup> the transition energy to the ground state is  $1.345 \pm 0.002$  MeV; and from the  $\gamma$ -ray energy, that to the excited state is  $0.227 \pm 0.005$  MeV. The neutrino energies for  $K$  capture,  $q_K$ , are obtained by subtracting the  $K$ -binding energy  $K_{ab}$ , of Cu ( $0.009$  MeV<sup>14</sup>) from the transition energies. Similarly,  $q_{L(1)}$  is obtained from  $L_{Iab}$  ( $0.001$  MeV). The  $(g_{L(1)}/g_K)^2$  values obtained from Brysk and Rose's paper<sup>7</sup> are multiplied by  $(q_{L(1)}/q_K)^2$  to get the two  $L/K$ -capture ratios  $(P_{L(1)}/P_K)$ , 0.0936 and 0.0994, where the uncertainty introduced by the energy limits of error in the last (excited state) ratio only amounts to  $\pm 0.2\%$ . Gleason,<sup>15</sup> determined that  $0.522 \pm 0.032$  of the electron capture transitions are to the excited state by

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\* Taken in part from the doctoral dissertation of Agustín G. Santos Ocampo.

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an x- $\gamma$  coincidence counting technique in which the  $\gamma$  efficiency of the crystal was determined in a separate experiment. This result does not depend on  $\omega_K$  or  $P_L/P_K$ . Weighting the two  $L_I/K$ -capture ratios by the branching ratio, the average  $P_{L(I)}/P_K$  is 0.0966 with an uncertainty due to the uncertainties in the energies and the branching ratio of only several tenths of a percent. Correcting this by the  $L_{II}/L_I$ -capture ratio<sup>7</sup> of  $6.8 \times 10^{-3}$ , the total average value of  $P_L/P_K$  is 0.097<sub>3</sub>. If the average branching ratio quoted by Strominger *et al.*<sup>13</sup> (44% to excited state) were used,  $P_L/P_K$  would be reduced by 0.4%. It is seen that the uncertainty in the theoretical  $P_L/P_K$  introduced by the uncertainties in neutrino energies and the branching ratio is much less than the approximately 4% uncertainty<sup>1</sup> in the theoretical results of Brysk and Rose.

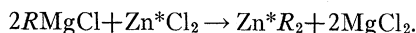
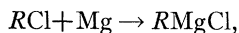
Neither the positrons nor the  $\gamma$  rays introduce experimental difficulties. The proportional counter is almost transparent to the essentially unconverted  $\gamma$  rays, and the Compton electrons will either deposit sufficient energy in the ring counters to close the anti-coincidence gate or merely cause a slight increase in the baseline of the spectrum. The same alternatives are true for the low-intensity (1.70%<sup>15</sup>) positron branch.

### APPARATUS

The apparatus has been described elsewhere.<sup>4</sup> Following the first determination at one atm to be reported, thicker wires of 9-mil stainless steel were used in place of 2-mil tungsten wires as cathodes in both center and ring counters. In addition, preliminary results<sup>16</sup> showed a longer delay time to be required for efficient blocking of the sample pulses, 2  $\mu$ sec being the value chosen.

### EXPERIMENTAL PROCEDURE

Two radioactive zinc dialkyls ( $Zn^*R_2$ ) were synthesized by the Grignard method:<sup>16,17</sup>



A small quantity of a solution of  $Zn^{65}Cl_2$  obtained from the Oak Ridge National Laboratory was evaporated on an Al planchet which was then added to a dropping funnel containing carrier  $ZnCl_2$  dissolved in ether. The solution was agitated vigorously and added dropwise to an ether solution of the Grignard reagent prepared from the appropriate alkyl chloride. Dibutyl ether, purified by two distillations through a 3-ft column packed with glass helices, the second from NaH, and diethyl ether were used as solvents in the synthesis of  $Zn^*(CH_3)_2$  and  $Zn^*(C_2H_5)_2$ , respectively. Each  $Zn^*R_2$  was separated from the solvent by distillation and

further purified by tube-to-tube distillation on a vacuum line.

Each time the counter was exposed to air, it was evacuated at 80°C to a residual pressure of 2  $\mu$ . Preliminary steps were taken to saturate the inner walls of the counter and premixing tank with inactive Zn ( $C_2H_5$ )<sub>2</sub> in an attempt to keep the contamination of  $Zn^{65}$  on the walls to a minimum. The counter filling procedure involved first vaporizing a trace of  $Zn^*R_2$  into a 50-liter mixing tank (corresponding to 0.02–0.05 mm Hg of  $Zn^*(CH_3)_2$  or 0.02–0.1 mm Hg of  $Zn^*(C_2H_5)_2$  in the counter). Then P-10 gas (9Ar/ $CH_4$ ) was added through a Ca purifier at 300°C and two dry ice-trichloroethylene baths and the mixture allowed to stand at least 24 h before expansion into the counter. It was hoped that this treatment would completely remove counter poisons such as oxygen and water which would decompose the  $Zn^*R_2$  and might broaden the spectrum at 4-atm P-10.<sup>4</sup> Actually, only a few complete spectra were obtained above 2-atm pressure as the counter usually went into continuous discharge before both peaks could be measured.

### RESULTS AND DISCUSSION

Typical  $K$ - and  $L$ -peak spectra at 2 atm of P-10 are shown in Figs. 1 and 2. The energy resolutions of the  $K$  and  $L$  peaks are 14 and 40%, respectively. After the background was subtracted, there remained activity between the  $L$  and  $K$  peak, presumed to be caused by degraded  $K$  pulses, with a height equal to 1.3% of the  $L$ -peak height. After extrapolation of this baseline, the baseline areas under each peak (channels 20–80) were typically found to be 2–3% and 4–5% of the  $K$  and  $L$  peaks, respectively. These baseline areas were subtracted from the background-corrected  $L$  and  $K$  peaks for the reason given in the previous study.<sup>4</sup>

The  $L/K$ -capture ratio was calculated from the

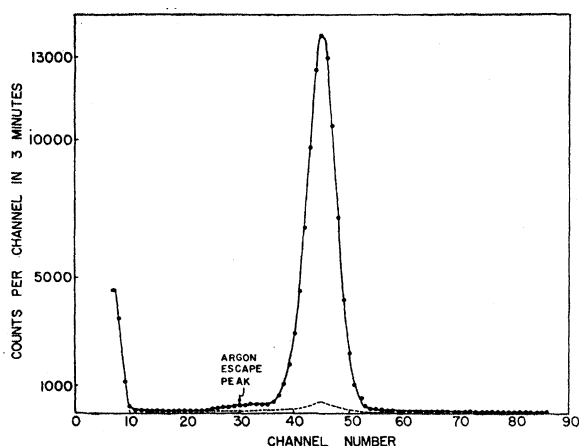


FIG. 1. The 9.0-keV  $K$ -peak spectrum of  $Zn^{65}$  in the center counter in anticoincidence with the ring counters. The background has not been subtracted and appears as a dashed line. The peak in the background is due to the contamination activity.

<sup>16</sup> A. G. Santos Ocampo, Doctoral dissertation, Purdue University, 1961 (unpublished).

<sup>17</sup> T. Kusama and D. Koike, J. Chem. Soc. Japan **72**, 871 (1951).

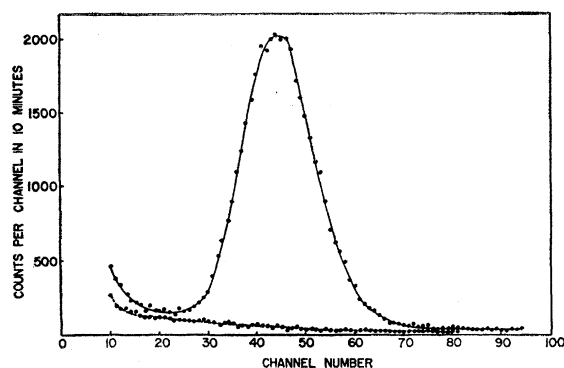


FIG. 2. The 1.10-keV  $L$ -peak spectrum of  $Zn^{65}$  in the center counter in anticoincidence with the ring counters. The background has not been subtracted and appears as a dashed line.

equation<sup>2</sup>

$$P_L/P_K = (N_L/N_K)[1 - (P_2 + P_3)\omega_K] - (P_1 + P_2 + P_3)\omega_K k, \quad (1)$$

where  $N_L/N_K$  is the ratio of the number of counts per unit time in the corrected  $L$  peak to that in the corrected  $K$  peak, and  $k$  is the fraction of  $K_\alpha$  x rays in the  $K$  series of Cu,<sup>18</sup> 0.88 (Table I). The  $P_i$ 's are the proba-

TABLE I.  $L/K$ -capture ratios of  $Zn^{65}$ .

Pressure (atm)	$P_1 \times 10^3$	$P_2 \times 10^3$	$P_3 \times 10^3$	$N_L/N_K$	$P_L/P_K$
1	9.5	0.8	41.8	0.1409	0.1205 <sup>a</sup>
				0.1386	0.1171 <sup>a</sup>
				0.1402	0.1187 <sup>a</sup>
2	5.9	2.3	3.1	0.1214	0.1172
				0.1236	0.1194
				0.1234	0.1192
				0.1242	0.1200
				0.1224	0.1202
3	4.2	1.6	0.3	0.1271	0.1255 <sup>b</sup>
4	3.2	1.2	0.0		
Average:					0.119 ± 0.007

<sup>a</sup> Not included in the average because of the large  $K$  x-ray escape corrections.

<sup>b</sup> Not included in the average because of the large deviation from the mean and the unusually large background correction. (The fraction of each total peak due to background was more than 50% larger than for any of the other 5 determinations.)

bilities for a  $K$  x ray to escape the center counter and either pass through one of the ends ( $P_1$ ), strike a cathode wire ( $P_2$ ), or pass between the cathode wires and not be absorbed in the ring counters ( $P_3$ ). These escape probabilities were estimated from reactor theory<sup>19</sup> with a few further assumptions.<sup>16,20</sup>

<sup>18</sup> A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1935), 2nd ed., p. 640.

<sup>19</sup> S. Glasstone, *Principles of Nuclear Reactor Engineering* (D. Van Nostrand and Company, Inc., Princeton, New Jersey, 1956), pp. 591–598, 628–629.

<sup>20</sup> Moler (R. B. Moler, Master of Science thesis, University of Arkansas, 1961 (unpublished)) used different equations for  $P_1$  and  $P_3$ . It is felt that  $P_L/P_K$  should be evaluated in a region where the escape correction is so small that  $P_L/P_K$  is rather independent of the values used for the escape probabilities, so it was decided not to devote space to the derivation and presentation of our equations for  $P_1$  and  $P_3$ .

The error in the average value of  $P_L/P_K$  has been assumed to be rather large because of the experimental difficulties (Appendix). The experimental ratio is 23% larger than the weighted mean of the theoretical ratios, 0.097, which was calculated above. This difference, which is considered significant, can be compared with an average positive discrepancy of 10% for the 5 nuclides in the first transition series noted above and the largest deviation obtained heretofore, 17% for cobalt-58.<sup>5</sup> Although the effect of the electron correlations on the  $L/K$ -capture ratio probably decreases rapidly with increasing atomic number,<sup>8</sup> it would still be desirable if an Odier and Daudel type calculation were made for a nuclide in this region to determine what fraction of the discrepancy is caused by this effect.

## APPENDIX

Two sources of error in the present work, which were not encountered in the  $Ar^{37}$  investigation, should be mentioned. In the first place, the anticoincidence gate was set at 3.2 keV. Therefore, about 25% of the 3.0-keV  $Ar$   $K$  x rays which escaped the center counter blocked the recording of the  $K$ -escape peak in the center counter (Fig. 1), and hence the  $N_L/N_K$  values are too large. The error was estimated to be no more than 1–1.5% at 2 atm either by the use of the number of counts in the escape peak or by assuming that each  $K$  capture resulted in the removal of a  $K$  electron from 3  $Ar$  molecules, the maximum energetically possible.

In addition, the amount of radioactivity in the counters decreased rather rapidly during their operation, the decrease in the center counter amounting to some 10–15% per 30 min. Because of this, the ratio of the integrated  $L$  peak and the average of the  $K$  peaks taken before and after the  $L$  peak was used for  $N_L/N_K$ . Although a stainless steel counter liner was used in place of an aluminum liner and  $Zn^*(CH_3)_2$ , in place of  $Zn^*(C_2H_5)_2$ , the rate of loss was not affected significantly. However, when the background was obtained after each run by evacuating the counter for several hours and refilling with P-10 to the original pressure, the amount of detectable contamination activity was much less than the former decrease in activity. With the exception of the 4-atm data, the ratio of the background in the center counter under each peak (including contamination) to the integrated peak activity ranged from 0.05–0.14. Since this ratio only increased about 0.01 per run at a given P-10 pressure, the background could be corrected for this increase with sufficient accuracy. The contamination activity, which could be removed by washing the stainless steel liner and steel spacers with dilute  $H_3PO_4$ , was presumed to be present as  $Zn^*$  or possibly  $Zn^*O$ . [The slow increase in the background was not surprising because of the similar results obtained by Townsend<sup>9</sup> who used  $Zn^*(C_2H_5)_2$ . One can only speculate on the cause of the rapid change in sample activity.<sup>16</sup>]

This decrease in activity during the run indicates

that the activity cannot be distributed both uniformly and only in the gas phase as assumed in the derivation of Eq. (1), and therefore indicates a possible error in the experimental ratio. However, it was noted that the  $L/K$ -capture ratio was independent (within  $\pm 4\%$ ) of variations in several experimental parameters, such as the pressure and type of the  $Zn^*R_2$ , the pressure of P-10, the type of liner in the counter, and the length of time the activity was in the counter, e.g., successive ratios

were found to be constant (to  $\pm 2\%$ ) in counting periods, which were about 0.7–2 h, in which 3–4  $K$  peaks alternating with 2–3  $L$  peaks were recorded. Therefore, some degree of confidence in the accuracy of the ratio is felt in spite of the varying activity.

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### $\beta$ - $\gamma$ Circular-Polarization Correlation in the Decay of $Mn^{52}$

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The  $\beta$ - $\gamma$  circular-polarization correlation in the decay of  $Mn^{52}$  has been measured by means of Compton forward scattering. The constant  $A$  was found to be  $A = 0.00 \pm 0.02$  which implies a value of  $x = -0.08 \pm 0.03$  for the ratio between Fermi and Gamow-Teller contributions to the decay (standard deviations). This result is in agreement with theoretical estimates.

#### I. INTRODUCTION

THE measurement of the circular polarization of gamma radiation following an allowed beta decay has been of great interest in the last few years because it directly indicates the ratio between Fermi and Gamow-Teller contributions to the decay.<sup>1-22</sup>

According to the isotopic-spin selection rule,<sup>23</sup> this ratio should be zero in all cases of practical interest as long as the isotopic spin itself is a good quantum number. The experimental results, however, are somewhat conflicting. Older experiments<sup>1-8,10,12,15</sup> seem to indicate large Fermi contributions (about maximum interference terms) in many cases, while most of the recent experiments<sup>13,16-18,21-22</sup> show smaller or even vanishing Fermi contributions for light- or medium-weight nuclei.

In the case of  $Mn^{52}$ , Boehm<sup>6</sup> first measured a large positive ratio between Fermi and Gamow-Teller contributions. Later Boehm<sup>22</sup> gave a smaller but still

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<sup>5</sup> F. Boehm and A. H. Wapstra, *Phys. Rev.* **109**, 456 (1958).

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<sup>11</sup> H. H. Forster and N. L. Sanders, *Nuclear Phys.* **15**, 683 (1960).

<sup>12</sup> V. M. Lobashov, V. A. Nazarenko, and L. I. Rusinov, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **40**, 10 (1961) [translation: *Soviet Phys.—JETP* **13**, 6 (1961)].

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<sup>17</sup> H. Daniel and M. Kuntze, *Z. Physik* **162**, 229 (1961).

<sup>18</sup> H. Daniel, M. Kuntze, and O. Mehling, *Z. Naturforsch.* **16a**, 1118 (1961).

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<sup>20</sup> S. D. Bloom, L. G. Mann, and J. A. Miskel, *Phys. Rev.* **125**, 2021 (1962).

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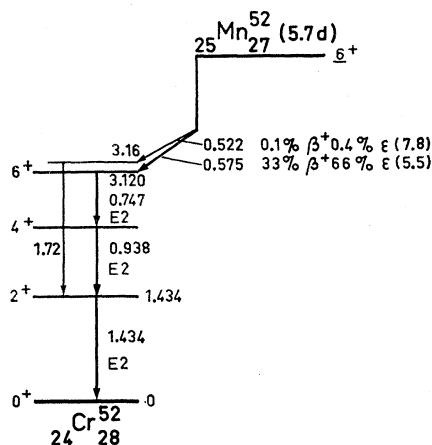


FIG. 1. Disintegration scheme of  $Mn^{52}$ .

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