

energy loss. At exactly forward, the two directions $\theta_R(\text{ad}; 0) = \frac{1}{2}\pi$ and $\theta_R(0) = 0$, are equivalent, as far as concerns the correlation pattern.

In what alpha-particle work there is available, the $\sin^2[2(\theta_\gamma - \theta_0)]$ form appears to be reasonably well satisfied, but it is not possible to distinguish between the adiabatic and plane-wave Born approximation predictions. More proton than alpha-particle work is available. In many cases the gamma distribution can be fitted with the form for spinless projectiles, and a comparison of the models is significant. The work of Sherr and Hornyak (16.6-Mev protons on C^{12}) clearly fits the adiabatic prediction for the symmetry angle

better than the plane-wave or distorted-wave Born predictions. However, the symmetry angles observed by Adams and Hintz (39.3-Mev protons on C^{12}) are intermediate between the adiabatic and the plane-wave Born predictions, somewhat favoring the latter, and are in good agreement with distorted-wave Born approximation computations. Experiments by Yoshiki (16.6-Mev protons on Mg^{24}) somewhat favor the adiabatic approximation.

More experiments conducted at forward scattering (angles of the order of $\Delta E/E$) would be highly desirable in distinguishing between the models; no such alpha-particle data are yet available.

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Decay of $\text{Hf}^{180m\frac{1}{2}}$ *

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The energies and relative intensities of the gamma radiation and the relative intensities of the conversion electrons following the decay of 5.5-hr Hf^{180m} have been measured using the curved-crystal gamma-ray spectrometer (recently calibrated for accurate intensity measurements), the ring-focused beta-ray spectrometer, and the semicircular spectrometer at the California Institute of Technology. The measured transition energies are: 57.54 ± 0.01 , 93.33 ± 0.02 , 215.25 ± 0.13 , 332.5 ± 0.3 , and 443.8 ± 0.6 kev. The energy levels deduced from these values are not entirely consistent with the two-parameter rotational formula. Conversion coefficients derived from the measurements were absolutely normalized using a method involving information available from the decay scheme. All of the 57.54-kev transition conversion coefficients are anomalously high if compared with the theoretical $E1$ coefficients. No admixture of $M2 + E3$ can account for the anomaly. The K conversion coefficients of the $E2$ transitions are all about 10% low with exception of the 93.33-kev transition. The L coefficients have a varying deviation, the maximum being 12%. The K conversion coefficient of the 501.3-kev transition has the value 0.037 ± 0.012 which is consistent with the theoretical $E3$ value of 0.040.

INTRODUCTION

THE energy levels of the Hf^{180} nucleus have for some time served as a classical example of a rotational excitation spectrum.¹ Above the 0^+ ground state four excited states with spin 2^+ , 4^+ , 6^+ , and 8^+ are known. The energies of these levels can be computed using a two-parameter formula of the form

$$E_I = (\hbar^2/2J)I(I+1) + BI^2(I+1)^2, \quad (1)$$

where J is the moment of inertia parallel to the symmetry axis, I the nuclear spin, and B is a constant taking into account the rotation-vibration interaction and other second order effects.¹ The Hf^{180m} γ -ray energies have now been measured with enough precision to pro-

vide a useful check of the validity of the two-parameter formula in this overdetermined set of data. It seemed to us worth while to undertake this precision measurement using the crystal diffraction spectrometer. The result to be described shows definite deviation from Eq. (1) for the 6^+ and 8^+ levels.

In addition, a precise evaluation of γ -ray and conversion-electron intensities seemed feasible and worth while in the Hf^{180} decay because of its simple cascade decay scheme. This evaluation results in precise absolute internal conversion coefficients for all $E2$ transitions of the rotational cascade as well as for the $E1$ and $E3$ transition from the 9^- intrinsic state (see Fig. 1).^{1a} Anomalies of the conversion coefficients of the 57.5-kev transition have been reported and discussed by Scharff-Goldhaber *et al.*² and Gvozdev and Rusinov.³

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¹ See, for example, S. A. Moszkowski in *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1957), Vol. 39, p. 485ff; and A. K. Kerman, in *Nuclear Reactions* (North-Holland Publishing Company, Amsterdam), p. 429.

^{1a} Note added in proof. From a recent experiment by M. Deusch and R. W. Bauer, Proc. Conf. Nuclear Structure, Kingston, 1960, p. 592, a spin assignment of 8^- follows for the 1142-kev state.

² G. Scharff-Goldhaber, M. McKeown, and J. W. Mihelich, Bull. Am. Phys. Soc. 1, 206 (1956).

³ V. S. Gvozdev and L. I. Rusinov, Doklady Akad. Nauk S.S.S.R. 112, 401 (1957); Soviet Phys.—Doklady 2, 35 (1957);

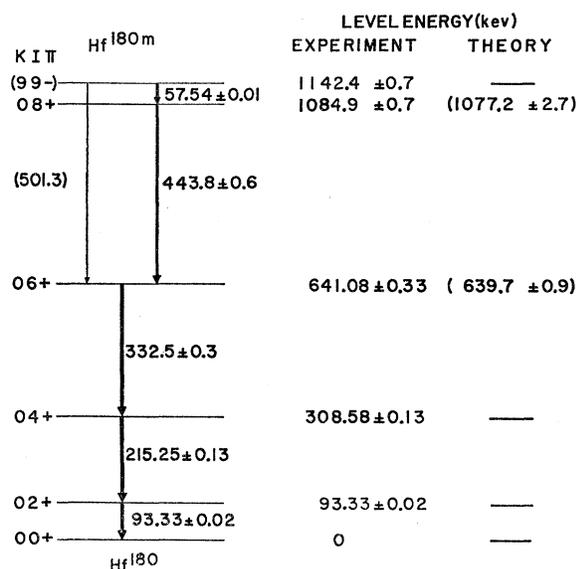


FIG. 1. Decay scheme of Hf^{180m} . The level energies which were deduced from the measured transition energies, are compared with theoretically predicted values based upon the two parameter formula fit to the 0+, 2+, and 4+ levels. The energy of the 501.3-keV crossover was deduced from the cascade gamma energies.

This transition is known to be $E1$ with a retardation factor of 10^{15} over the single-particle estimate.

INSTRUMENTS AND SOURCES

The 5.5-hour isomer Hf^{180m} was produced by neutron irradiation of HfO_2 enriched in the isotope 179. By using samples of high enrichment (83.6% Hf^{179}), recently made available through the Stable Isotope Division of the Oak Ridge National Laboratory, a contaminant activity due to radioactive Hf^{181} , produced by neutron capture of Hf^{180} , was minimized. Sources were irradiated in the Materials Testing Reactor at Arco, Idaho in a neutron flux of $2 \times 10^{14}/\text{cm}^2$ for approximately six hours.

Gamma-ray energies and intensities were measured with the two-meter curved-crystal spectrometer. This instrument has previously been described.⁴ Under the present conditions the energy resolution is $\Delta E/E = 2.3 \times 10^{-5}E$, where E is the gamma-ray energy in keV and ΔE is the full width of the line profile at half-maximum. The spectrometer was recently calibrated to permit precise relative intensity measurements.⁵ Corrections due to absorption in the air path, absorption in the bent quartz crystal, absorption in the aluminum window of the NaI(Tl)-crystal container, photopeak efficiency of the detector, and reflectivity of the quartz crystal were applied. Further corrections due to self-absorption in the source and absorption in the source container were

and V. S. Gvozdev, L. I. Rusinov, Yu. I. Filimonov, and Yu. L. Khazov, *Nuclear Phys.* **6**, 561 (1958).

⁴ J. W. M. DuMond, *Ergeb. exakt. Naturw.* **38**, 232-301 (1955).

⁵ W. F. Edwards, Ph.D. thesis, California Institute of Technology, 1960 (unpublished).

considered. By far the most important correction is due to the energy dependence of the reflectivity of the curved quartz-crystal. As a secondary result from the present study, when combined with the results of other experiments, this energy dependence was found to be $E^{-1.987 \pm 0.022}$ as will be discussed below.

The gamma-ray source material was enclosed in a fused quartz capillary tube of internal diameter 0.009-in., external diameter 0.050 in., and height 1 in. The ends of the capillary tube containing the material were fused together, forming a completely enclosed "line" source.

The homogeneous-field ring-focusing beta-ray spectrometer and the semicircular spectrometer were used to measure the relative intensities of the conversion electrons. These instruments are described in the literature.^{6,7} The ring-focusing spectrometer is used in the measurements of electrons having energies greater than 25 keV. Its detector is a Geiger counter having a mica window of thickness 0.9 mg/cm². The semicircular spectrometer was employed to measure beta rays having energies between 2 keV and 120 keV. The Geiger counter detector had a formvar window of 10-30 $\mu\text{g}/\text{cm}^2$ thickness made by a standard process.⁸

Sources for the two spectrometers were prepared by vacuum evaporation of the activated material onto aluminum backings. The ring-focusing spectrometer was operated with a resolution of 0.7%. The sources were disks, 3 mm in diameter. The semicircular spectrometer sources were 1.5 mm \times 2.5 cm rectangles matched for a 1% momentum resolution.

RESULTS

Gamma-ray energies which were obtained by matching the profiles of the gamma lines resulting from reflection from the opposite sides of the crystal planes, are given in Table I. The error assignments vary from 1/20 of the width of the line for strong lines to 1/7 of the width in the case of the weak 443.8-keV line. The energy of the 501-keV line was not measured. Gamma-ray relative intensities were found by setting the diffraction spectrometer on the maximum position of the line profiles and recording the pulse-height spectrum of the NaI detector with a 100-channel analyzer. The relative intensities were taken to be equal to the areas of the photopeaks after being corrected for the effects previously mentioned. The gamma-ray relative intensities are given in Table I.

Relative electron intensities were measured by determining the counting rate as a function of the magnetic rigidity, $B\rho$. The relative intensity of a line is then equal to the area of the line profile divided by $B\rho$. The only

⁶ J. W. M. DuMond, *Ann. Phys.* **2**, 283 (1957).

⁷ H. E. Henrikson, California Institute of Technology Report USAEC No.—24, 1956 (unpublished).

⁸ H. Slatis, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 52.

correction that was necessary to be applied was due to the dead-time of the Geiger counter. This correction never contributed more than 4% to the intensity of the line.

The ring-focusing spectrometer was used to measure the 93.33-keV L -conversion lines and all lines with higher energies. The semicircular spectrometer was used to measure the 93.33-keV M -conversion lines and all lines with lower energy. As an illustration the 57.54-keV L -conversion lines and the 93.33-keV K -conversion line are shown in Fig. 2.

ROTATIONAL ENERGY LEVELS

From the transition energies measured with the curved-crystal spectrometer the level energies given in Fig. 1 were deduced. The energy values of the first two excited levels were used to determine the parameters $\hbar^2/2J$ and B of Eq. (1). The following numbers were found: $\hbar^2/2J = (15.609 \pm 0.006)$ keV and $B = -(0.0090 + 0.0005)$ keV. The energies of the 6+ and 8+ levels can now be predicted from these values of the parameters. By comparing the predicted values and the experimental data for the 6+ and 8+ level (Fig. 1), it can be seen that the former are lower than the experimental values and there is no overlap of the errors.

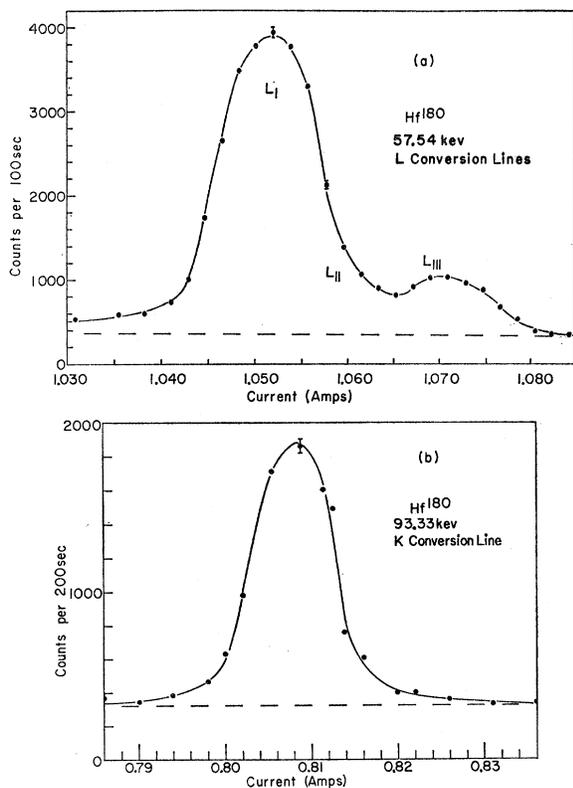


FIG. 2. Conversion electron lines measured using the semicircular spectrometer. (a) shows the 57.54-keV transition L lines, and (b) shows the K line of the 93.33-keV transition.

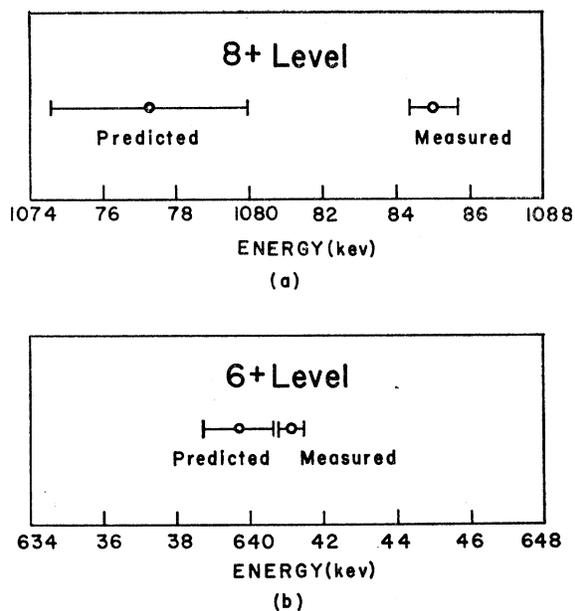


FIG. 3. Comparison of the experimental values of the energies predicted by the two parameter formula for the 6+ and 8+ levels in Hf^{180} . The uncertainties represent 90% confidence limits.

Figure 3 shows the relationship between the values and the errors on the predicted and the experimental energies. One should remember that the reported energy uncertainty for lines measured with the curved-crystal spectrometer is not a standard deviation, but corresponds to about 90% confidence. Thus the probability of the deviation of the measured energy of the 6+ level being statistical is approximately 0.03 while that for the 8+ level is smaller yet. A significant fact is that the deviations are increasing with energy. We must conclude, that the two-parameter rotational formula does not explain the experimental observations.⁹

NORMALIZATION OF CONVERSION COEFFICIENTS

The experimental relative conversion coefficients listed in Table I were found by dividing the intensity of each conversion line by the corresponding gamma-ray intensity. These relative coefficients were then sub-

⁹ The above procedure for fitting Eq. (1) to the experimental data has visual advantages but is not a proper least-squares adjustment. [For a discussion of the least-squares method see E. R. Cohen, K. M. Crowe, and J. W. M. DuMond, *Fundamental Constants of Physics* (Interscience Publishers, Inc., New York, 1957).] A least-squares adjustment was performed with the result that $\hbar^2/2J = (15.596 \pm 0.004)$ keV and $B = -(0.00745 \pm 0.00014)$ keV. Furthermore the ratio of the external error (which is the standard deviation based upon the spread of the observations) and internal error (which is the standard deviation reflecting the accuracy of the original measurements) is 5.9. If the data were entirely self-consistent this ratio would be equal to 1. Since the method of assigning errors to the curved-crystal spectrometer energy measurements has been carefully checked in the past, there is little reason to suspect that the internal error is faulty. This implies that the spread of the observations is highly excessive. The probability of this being a random deviation is much less than 0.01.

jected to an absolute normalization in the following manner:

The decay fraction of a transition is the percentage of the total decay rate contributed by a particular electromagnetic transition and is proportional to $(1+\alpha_i)I_\gamma$, where α_i is the total conversion coefficient of the transition, and I_γ is the relative intensity of the gamma ray. If no primary beta branching exists as in the case of Hf^{180} then the sum of the electromagnetic decay fractions of transitions populating the level must clearly be equal to the sum of those depopulating the level, thus yielding for each level an equation containing the relative conversion electron intensities, the relative gamma-ray intensities, and one absolute conversion coefficient only. Thus, with one level to which this decay scheme method can be applied, the conversion coefficients may be absolutely determined.

Hf^{180m} has four levels for which such equations can be written: the 2+, 4+, 6+, and 8+ levels. The gamma-ray intensity of the 501-keV transition was not, however, determined with sufficient accuracy. As a result the 6+ level equation contained two unknown quantities. In addition, the uncertainties of the 443.8-keV transition intensities were quite large. For these

reasons this equation was rejected leaving three equations in one unknown, the unknown being the conversion coefficient of the 93.33-keV transition.

The equations were thus twice overdetermined. Advantage was taken of this overdetermination to check the intensity calibration of the curved-crystal spectrometer. The gamma-ray relative intensities had been corrected assuming a $E^{-2.000}$ dependence of the curved-crystal reflection coefficient. An unknown, x , was then introduced representing the deviation of the actual exponent from the one assumed. Thus, through the gamma intensities, x appeared as a second unknown in the three level equations. Now, in another experiment a value for x had been determined. This was introduced as a fourth equation, $x=0.000\pm 0.032$, giving finally four equations in two unknowns. A least squares adjustment was then made, taking into account the correlations between parameters of the input data. The resulting value for the total conversion coefficient of the 93.33-keV transition is 5.14 ± 0.24 . The internal error is given. The ratio of the external error and internal error is 0.8 which indicates very good consistency of the data. The adjusted value for x was -0.013 ± 0.022 , indicating a $E^{-1.987\pm 0.022}$ reflectivity law.

TABLE I. Data for transitions in Hf^{180m} .

Gamma-ray energy (keV) ^a	Gamma-ray intensity (relative units)	Conversion line	Conversion electron intensities (relative units)	Absolute, by present experiment ^b	Conversion coefficients			
					Experiment by Gvozdev and Rusinov ^c		Theoretical ^d	
57.54±0.01	291± 7	L_I+L_{II}	0.248 ±0.014	0.458 ±0.036	0.33 ±0.10		$E1$	$M2$
		L_{III}	0.045 ±0.006	0.084 ±0.012			0.163	69
		L_{total}	0.294 ±0.012	0.543 ±0.036			0.062	22
		M	0.072 ±0.007	0.134 ±0.015			0.225	91
		$N+O$	0.012 ±0.002	0.023 ±0.004			0.086 ^e	43
93.33±0.02	100± 2	K	0.205 ±0.012	1.10 ±0.09	1.3 ±0.4		$E2$	
		L_I+L_{II}	0.343 ±0.017	1.85 ±0.13			1.03	
		L_{III}	0.239 ±0.013	1.29 ±0.09			1.46	
		L_{total}	0.582 ±0.017	3.13 ±0.19			1.32	
		$M+N+\dots$	0.169 ±0.012	0.909 ±0.08			2.80	
215.25±0.13	500±14	K	0.114 ±0.005	0.123 ±0.009	0.15 ±0.05		$E2$	
		L_{total}	0.072 ±0.006	0.077 ±0.007			0.138	
332.5 ±0.3	567±24	K	0.0400 ±0.0016	0.038 ±0.003	0.055 ±0.014		$E2$	
		L_{total}	0.0154 ±0.0012	0.0146±0.0015			0.042	0.0130
443.8 ±0.6	491±26	K	0.0173 ±0.0010	0.0189±0.0017	0.026 ±0.007		$E2$	
		L_{total}	0.0040 ±0.0005	0.0044±0.0007			0.020	
		$M+N+\dots$	0.00141±0.00028	0.0015±0.0003			0.0063±0.0016	0.0049
		total	0.0227 ±0.0012	0.0249±0.0022				
501 ^f	102±31 ^g	K	0.0070 ±0.0009	0.0370±0.012	0.035 ±0.014		$E3$	$M4$
							0.040	0.77

^a While all other errors reported in this paper represent standard deviations the energy errors correspond to ~90% confidence limits.

^b The normalization of these conversion coefficients was fixed by $\alpha(93)$ whose absolute value was determined as described in the text.

^c See reference 3.

^d See reference 10.

^e Not corrected for screening.

^f The energy of the 501-keV line was not measured.

^g This value was not measured directly but was deduced from the feeding and bleeding of levels as discussed in the text.

CONVERSION COEFFICIENTS

Table I gives the absolute conversion coefficients, comparing them with the values determined by Gvozdev and Rusinov³ and the theoretical values of Rose.¹⁰ The 57.54-keV *L*-conversion coefficient anomaly is confirmed. Scharff-Goldhaber *et al.*² report the ratios $L_I/L_{II}/L_{III}=5:0.5:1$ which gives $(L_I+L_{II})/L_{III}=5.5$. The present work gives the same value for this ratio, 5.5 ± 0.8 . Rose's theoretical value is $(L_I+L_{II})/L_{III}=2.63$. Gvozdev and Rusinov report a value for the last ratio of approximately 4. Figure 2(a) shows the 57.54 *L*-conversion lines. From the figure, it can be seen that L_I gives the main contribution to the composite line in agreement with Scharff-Goldhaber. The value obtained for the total *L*-conversion coefficient is 0.54 ± 0.04 , while Scharff-Goldhaber *et al.*, give 0.4 and Gvozdev and Rusinov give 0.33 ± 0.10 . The curved-crystal spectrometer made it possible to resolve the 57.54-keV line from the Hf *K* x rays, so the gamma-ray intensity was subject to a very small error only.

M2 admixture will not explain the anomaly. The amount of *M2* necessary to bring agreement with α_{LIII} is $(0.10\pm 0.06)\%$; however, this would only raise the theoretical value for $\alpha_{LI}+\alpha_{LII}$ to 0.24 ± 0.04 , whereas the experimental value for this last quantity is 0.44 ± 0.03 .

The present result is in general agreement with the results of Asaro *et al.*, on the anomalous conversion coefficients of retarded, low-energy, *E1* transitions.¹¹ These authors found a definite correlation between the *L* shell conversion coefficient anomaly and gamma-ray retardation in odd-*A* isotopes in the trans-lead region. They also report that the L_{III} subshell is not anomalous. In the present case L_{III} is close to the theoretical value while L_I+L_{II} is three times larger than the theoretical value.

The 57.54-keV *M* coefficient is approximately 60% higher than the theoretical *E1* value. The screening effect, which was neglected in Rose's calculation of the *M* coefficients, would lower the theoretical value.¹² Furthermore, no ratio of *M2*+*E3* admixture can satisfactorily explain the anomaly.

It was mentioned that the gamma intensity of the

TABLE II. Subshell intensity ratios of conversion electrons for transitions in Hf^{180m}.

Gamma-ray energy (keV)	Identification	Measured value	Value by Gvozdev <i>et al.</i> ^a	Theoretical value ^a
57.54	$(L_I+L_{II})/L_{III}$	5.5 ± 0.8	~ 4	<i>E1</i> 2.63
	L/M	4.1 ± 0.4	...	<i>M2</i> 3.13
	$L/(N+0\cdots)$	24 ± 4	...	2.62 2.12
93.33	<i>K/L</i>	0.352 ± 0.025	0.48 ± 0.15	<i>E2</i> 0.368
	$(L_I+L_{II})/L_{III}$	1.44 ± 0.10	1.4 ± 0.3	1.11
	$L/(M+N\cdots)$	3.44 ± 0.26
215.25	<i>K/L</i>	1.59 ± 0.14	...	<i>E2</i> 2.08
332.5	<i>K/L</i>	2.60 ± 0.23	...	<i>E2</i> 3.20
443.8	<i>K/L</i>	4.3 ± 0.6	4.1 ± 1.3	<i>E2</i> 4.1

^a See reference 3.
^b See reference 10.

501-keV transition was not determined in this experiment; for this reason the 6+ level equation, which involves this quantity was not used in the determination of the absolute value of the 93.33-keV conversion coefficient. After the determination of α (93.33), however, this equation was used to deduce values for the total conversion coefficient and gamma-ray intensity of the 501-keV transition. These values are summarized in Table I. An *E3* assignment brings agreement between experimental and theoretical conversion coefficients within the rather large error.

The following comment applies to the *E2* transitions. With the exception of the 93-keV line the *K*-conversion coefficients of all *E2* lines show a slight trend toward lower values if compared with the theoretical coefficients in contrast with the reported³ trend toward higher values. The total *L*-coefficients seem to be somewhat higher than the theoretical values. This variation of *K*-shell and *L*-shell coefficients shows up clearly in the *K/L* ratios given in Table II. The subshell ratios are in agreement with those reported by Gvozdev *et al.*³

ACKNOWLEDGMENTS

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¹⁰ M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

¹¹ F. Asaro, F. S. Stephens, J. M. Hollander, and I. Perlman, *Phys. Rev.* **117**, 492 (1960).

¹² M. E. Rose (privately circulated tables of conversion coefficients).