Internal-conversion studies in ^{177}Hf from the decay of ^{177}Lu

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The K-shell and L-subshell internal-conversion coefficients of the retarded transitions in ^{177}Hf populated in the decay of the 6.8-day ^{177}Lu have been measured using a high-resolution double-focusing electron spectrometer and a 20-cm^3 Ge(Li) detector. The K conversion coefficients of the 71.6- and 113-keV transitions have been determined by γ-γ and γ-x-ray coincidence measurements using a 1-cm^3 Ge(Li) detector having a thin window and a scintillation detector. The γ-γ, γ-e^K, and e^K-γ angular correlations of the 208.3-113-keV cascade have been measured using a single gap electron spectrometer, NaI(Tl) γ-ray spectrometers, and a Ge(Li) detector. The particle parameters for the K conversion electrons of the transitions have been evaluated to be: $b_2^K(208.3) = -1.54 \pm 0.06$ and $b_2^K(113) = -0.20 \pm 0.02$. The 208.3-113-keV γ-γ angular correlation as measured using a $LuCl_3$ source in solution gave $W(\theta) = 1 - (0.192 \pm 0.007) P_2(\cos\theta)$. The quadrupole admixtures and the penetration parameters of the transitions were evaluated by least-squares analysis of the conversion coefficients and particle parameters. The best solutions are $Q = (3.1 \pm 1.6)\%$, $\lambda_1 = -19.2 \pm 1.2$, $\lambda_2 = 250 \pm 240$ for the 321.3-keV transition; $Q < 0.2\%$, $\lambda_1 = -0.8 \pm 0.9$, $\lambda_2 = 0$ for the 208.3-keV transition; $Q = (0.03 \pm 0.02)\%$, $\lambda_1 = 1.3 \pm 0.8$ for the 71.6-keV transition; $Q = (94.1 \pm 0.7)\%$, $\lambda' = 5.6 \pm 8.4$ for the 113-keV transition, and $Q = (95 \pm 5)\%$ for the 136.7-keV transition. From these values and the known lifetimes of the levels the γ-ray matrix elements and the penetration matrix elements have been calculated and compared with theoretical estimates based on Nilsson's model.

RADIOACTIVITY ^{177}Lu; measured I_γ, I_{ce}, xγ and $\gamma\gamma$ coin, $\gamma\gamma(\theta)$, ce $\gamma(\theta)$. ^{177}Hf, deduced ICC, b_2^K, δ, penetration parameters. Double focusing and single gap e spectrometers, Ge(Li) detectors. Enriched target.

I. INTRODUCTION

The low-lying levels of ^{177}Hf populated in the decay of the 6.8-day ^{177}Lu have been extensively studied.[1-3] The decay scheme[4] is shown in Fig. 1. The 71.6-, 208.3-, and 321.3-keV transitions are characterized as $K = \frac{9}{2}^+[624]\uparrow \rightarrow K = \frac{7}{2}^-[514]\downarrow$ interband $E1$ transitions. These are highly retarded transitions with retardation factors $>10^4$. Since these transitions are not K forbidden, penetration effects could be expected[5,6] in their internal conversion. The $M1$ component of the 113-keV intraband $\frac{9}{2}^- \rightarrow \frac{7}{2}^-$ transition is also highly retarded. The retardation in this case is due to accidental cancellation of the collective and intrinsic gyromagnetic ratios. Observable penetration effects are expected[7] in this transition also.

Bashandy, El-Sayad, and El-Aassar[8] have determined the K conversion coefficient of the 321.3-keV transition which was found to be approximately 10 times larger than the theoretical value for $E1$. They have attributed this discrepancy to penetration effects. Hager and Seltzer[9] have measured the conversion coefficients and L-subshell ratios of the 321.3-, 208.3-, and 71.6-keV transitions and have analyzed for the penetration parameters. The value of the K conversion coefficient of the 321.3-keV transition reported by them is not in agreement with previous measurements.[1,8] The quadrupole admixture and the penetration parameters could be evaluated more unambiguously from the absolute K, L_1, L_{11}, and L_{111} conversion coefficients. Since there are discrepancies in the existing measurements it was found necessary to remeasure the conversion coefficients. Some of the conversion coefficients had to be newly measured.

The K conversion coefficient of the 113-keV transition has been measured by Marmier and Boehm[1] and by Alexander, Boehm, and Kankeleit.[2] The L-subshell ratios of this transition have been measured by Alexander, Boehm, and Kankeleit,[2] Novakov and Hollander,[10] and by Hogberg *et al.*[11] However, the penetration parameter for this transition could not be evaluated unambiguously because of the uncertainty in the mixing ratio. Furthermore, the results of measurements by different groups[2,10,11] are not in agreement with each other.

Measurements of e^--γ, γ-e^-, and γ-γ angular correlations of the 208.3-113-keV cascade have been made by a number of workers.[12-20] However,

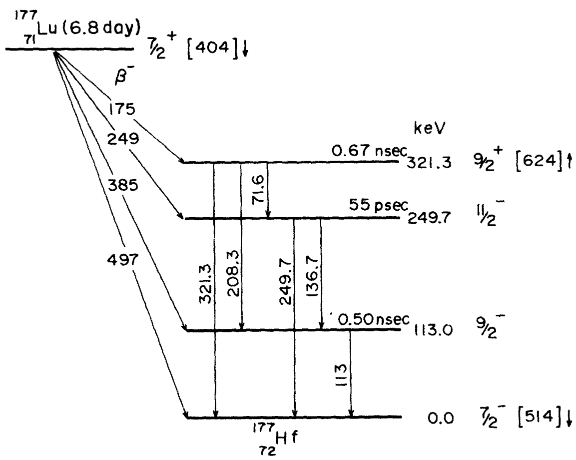

FIG. 1. The decay scheme of ^{177}Lu.

it has not been possible to come to a definite conclusion about the multipole mixing amplitudes and the penetration effects in the 208.3- and 113-keV transitions because of considerable disagreement among the results of measurements.[12-20] Thun *et al.*[12] have determined the particle parameter for K electrons for the 208.3- and 113-keV transitions from e^K-γ, γ-γ, and γ-e^K angular correlations. Their value for the particle parameter for the 113-keV transition is in disagreement with later measurements[13-15] and also with the theoretical value without penetration effects.[21] There is also disagreement among the values of the γ-γ angular correlation coefficient measured by various workers. The analysis of the e^--γ, γ-e^-, and γ-γ angular correlation measurements of the 208.3-113-keV cascade is complicated because both the transitions could have quadrupole admixture and penetration effects. It is preferable to evaluate the mixing ratios and penetration parameters for both these transitions simultaneously from the absolute values of the conversion coefficients, particle parameters, and γ-γ angular correlation coefficients.

In the present work the absolute K-shell and L-subshell conversion coefficients of the 113-, 136.7-, 71.6-, 208.3-, and 321.3-keV transitions have been determined. The particle parameters for K-conversion electrons of the 208.3- and 113-keV transitions and the 208.3-113-keV γ-γ angular correlations have been measured. The penetration parameters λ_1 and λ_2 and the $M2$ admixtures Q of the 321.3- and 71.6-keV transitions have been deduced from a least-squares fit of the measured conversion coefficients. The penetration parameters and the amplitude mixing ratios of the 208.3- and 113-keV transitions have been extracted simultaneously by least-squares analysis of the conversion coefficients, particle parameters, and the γ-γ angular correlation coefficient measured using a liquid source. The penetration parameters deduced from experiments are compared with the theoretical estimates based on Nilsson's model. Preliminary reports of our measurements were presented earlier.[22] When the present measurements were nearly complete the results of measurements of the internal conversion coefficients of the 71.6-, 208.3-, and 321.3-keV transitions by Grigorev, Kaminkar, and Sergenkov[23] were published. The present results are in agreement with their values.

II. EXPERIMENTAL

A. Source preparation

Sources of ^{177}Lu were made by irradiating 72% enriched ^{176}Lu in the form of Lu_2O_3 in the CIRUS reactor at the Bhabha Atomic Research Centre, Trombay. For the conversion electron measurements the enriched Lu_2O_3 was vacuum evaporated onto a 1-mg/cm^2 pure-aluminum foil. Pieces of this foil, 1 mm × 15 mm in size, were irradiated for one week in the reactor. A piece of this foil was stuck onto a 0.8-mg/cm^2 aluminized Mylar film and used for the conversion electron measurements with the double focusing spectrometer. A piece of the same sample was used for the γ-ray measurements. Another piece <2 mm in length was used for e^--γ and γ-γ angular correlation measurements to determine the particle parameters. The γ-γ angular correlation was also measured with the source in the form of a dilute solution of $LuCl_3$ in dilute HCl. The sources used for the e^--γ angular correlation and conversion electron measurements were thin enough (<5 μg/cm^2) so that self-absorption and scattering corrections were negligible.

B. γ-ray measurements

γ-ray spectra of ^{177}Lu decay were measured using a 20-cm^3 Ge(Li) detector having a resolution of ~3.5 keV at 662 keV. The relative photopeak efficiencies as a function of γ-ray energy for the Ge(Li) detector were determined by measuring the γ spectra of ^{160}Tb, ^{152}Eu, and ^{169}Yb which have well-known relative intensities.[4,24] The relative γ-ray intensities in the decay of ^{177}Lu obtained from the analysis of the spectrum in the present work are shown in Table I.

C. Internal-conversion-electron measurements

The internal-conversion-electron spectra were recorded using an iron yoke double-focusing electron spectrometer with a momentum resolution of ~0.1%. The spectra were analyzed in the manner described in Ref. 24. The decay of each conversion line was followed for several half-lives of ^{177}Lu to ensure that contributions of any possible impurities were negligible. Corrections for the background due to the β continuum in the decay of ^{177}Lu and for the absorption of electrons by the ~1 mg/cm^2 counter window were applied.[24] The sloping β background under the L conversion spectra was determined by an iterative procedure.[24] The relative intensities of the conversion lines were calculated and averaged over several runs. The measured relative electron intensities are shown in Table I. The absolute conversion coefficients were determined from the relative intensities of the conversion electrons and γ rays using the K conversion coefficient of the 307.7-keV transition[24] in ^{169}Tm as standard. The experimental values of the conversion coefficients along with the theoretical values without penetration effects[21] are listed in Table II.

D. γ-γ and γ-x-ray coincidence measurements

Because of the low energy of the K electrons of the 71.6- and 113-keV transitions their relative intensities could not be accurately determined by the double-focusing spectrometer. Hence the K conversion coefficients of these transitions were determined by γ-γ and γ–x-ray coincidence measurements. A 1-cm^3 Ge(Li) detector with a thin window having a resolution of ~3.2 keV at 113 keV and a 3.8-cm-diam × 5.0-cm-thick NaI(Tl) crystal mounted on an RCA 6810A photomultiplier and a time-to-pulse-height converter were used. The full prompt peak in the time-to-pulse-height converter spectrum in the required energy region was gated by a single-channel analyzer. Another single-channel analyzer was used to gate either the 208.3-keV or the 249.7-keV γ ray of ^{177}Lu. The outputs of the two single-channel analyzers were put in slow coincidence. The spectrum in the Ge(Li) detector gated by the slow coincidence output was scanned in a 400-channel analyzer. The over-all resolving time of the coincidence system as measured by observing the chance coincidence was $2\tau = 150$ nsec. A weak source of ^{177}Lu was used in order to keep the chance coincidence low. The angle between the two detectors was kept as 55° so that angular correlation effects could be neglected. An anti-Compton shield was used between the detectors to avoid spurious coincidences due to scattering.

The observed spectra in coincidence with the 208.3- and the 249.7-keV γ rays are shown in Fig. 2. The observed coincidences were corrected for chance coincidences which were <0.5% for coinci-

TABLE I. Relative intensities of γ rays and internal-conversion electrons in the decay of ^{177}Lu. Intensities of γ rays and the conversion electrons are normalized separately.

E_γ (keV)	I_γ	I_{e_K}	$I_{e_{L1}}$	$I_{e_{L11}}$	$I_{e_{L111}}$
71.6	1.5±0.1	···	2.9±0.1	1.2±0.2	1.1±0.2
113.0	60±5	···	119±6	720±22	627±19
136.7	0.52±0.05	5.5±0.7	0.9±0.2	3.8±0.4	3.4±0.5
208.3	100	100	13.7±0.6	2.4±0.2	2.2±0.2
249.7	1.9±0.2	4.1±0.2	···	···	···
321.3	2.0±0.2	4.4±0.3	0.74±0.07	0.15±0.03	0.015±0.010

TABLE II. Absolute K-shell and L-subshell conversion coefficients.

	Shell	Experiment	Theory (Ref. 21)	
(a) 71.6 keV	K	0.90±0.11	0.72	($E1$)
			92.0	($M2$)
	L_1	0.087±0.010	0.071	($E1$)
			24.3	($M2$)
	L_{11}	0.038±0.008	0.026	($E1$)
			2.58	($M2$)
	L_{111}	0.033±0.007	0.031	($E1$)
			7.30	($M2$)
(b) 208.3 keV	K	0.046±0.004	0.046	($E1$)
			2.05	($M2$)
	L_1	0.0063±0.0006	0.0053	($E1$)
			0.38	($M2$)
	L_{11}	0.001 10±0.000 12	0.000 92	($E1$)
			0.049	($M2$)
	L_{111}	0.001 00±0.000 12	0.000 90	($E1$)
			0.039	($M2$)
(c) 321.3 keV	K	0.102±0.013	0.0155	($E1$)
			0.49	($M2$)
	L_1	0.017±0.0024	0.0018	($E1$)
			0.081	($M2$)
	L_{11}	0.0034±0.0008	0.000 24	($E1$)
			0.010	($M2$)
	L_{111}	0.000 35±0.000 25	0.000 22	($E1$)
			0.0049	($M2$)
(d) 249.7 keV	K	0.101±0.009	0.26	($M1$)
			0.094	($E2$)
(e) 113 keV	K	0.78±0.05	2.4	($M1$)
			0.74	($E2$)
	L_1	0.091±0.011	0.345	($M1$)
			0.076	($E2$)
	L_{11}	0.55±0.05	0.031	($M1$)
			0.58	($E2$)
	L_{111}	0.48±0.04	0.0038	($M1$)
			0.48	($E2$)
(f) 136.7 keV	K	0.49±0.08	1.45	($M1$)
			0.48	($E2$)
	L_1	0.08±0.02	0.20	($M1$)
			0.05	($E2$)
	L_{11}	0.33±0.05	0.019	($M1$)
			0.24	($E2$)
	L_{111}	0.30±0.05	0.0020	($M1$)
			0.195	($E2$)

dence with 208.3-keV and <2% for coincidence with 249.7-keV γ rays. A small peak at 113 keV in the coincidence spectrum with the 249.7-keV γ ray in gate (see Fig. 2) is due to the tail of the 208.3-keV γ ray accepted in the gate. A correction for this contribution was applied using the observed coincidence spectrum with the 208.3-keV γ ray. Similarly a small correction of the coincidence with the Compton of the 249.7-keV γ ray accepted in the 208.3-keV gate was applied in the coincidence with the 208.3-keV γ ray. The relative intensities of the K_α, K_β, and γ-ray peaks were obtained from the analysis of the coincidence spectra. The relative photopeak efficiencies of the Ge(Li) detector as a function of energy under the same conditions were determined by the analysis of the spectra of ^{169}Yb, ^{203}Hg, ^{170}Tm, ^{133}Ba, and ^{57}Co. The conversion coefficient was calculated from the relation

$$\alpha_K = \frac{(N_\alpha/\epsilon_\alpha) + (N_\beta/\epsilon_\beta)}{\omega_K(N_\gamma/\epsilon_\gamma)}, \quad (1)$$

where N_α, N_β, and N_γ are the total counts under the K_α, K_β, and γ-ray peaks in the coincidence spectrum, ϵ_α, ϵ_β, and ϵ_γ are the relative photoefficiencies, and ω_K in the K fluorescence yield[25] of Hf.

E. e-γ angular correlations

The $e^K(208.3)$-$\gamma(113)$ and $\gamma(208.3)$-$e^K(113)$ angular correlations were measured using a single gap β-ray spectrometer[26] for detecting the conversion electrons. The γ rays were detected in a 4.4-cm-diam × 5-cm-thick NaI(Tl) detector kept at a distance of 7 cm from the source. The coincidence setup had a resolving time $2\tau = 60$ nsec. The e^--γ coincidences were measured at angles 105, 130, 155, 180, 205, 230, and 255°. The contribution to the β background was obtained by measuring coincidences after adjusting the spectrometer current to focus the β continuum just off the conversion-electron line. This contribution was found to be isotropic within experimental errors and was 12% of the gross coincidences at 105° for $\gamma(208.3)$-$e^K(113)$-keV cascade and 38% for $e^K(208.3)$-$\gamma(113)$-keV cascade. The e^K-γ angular correlations cor-

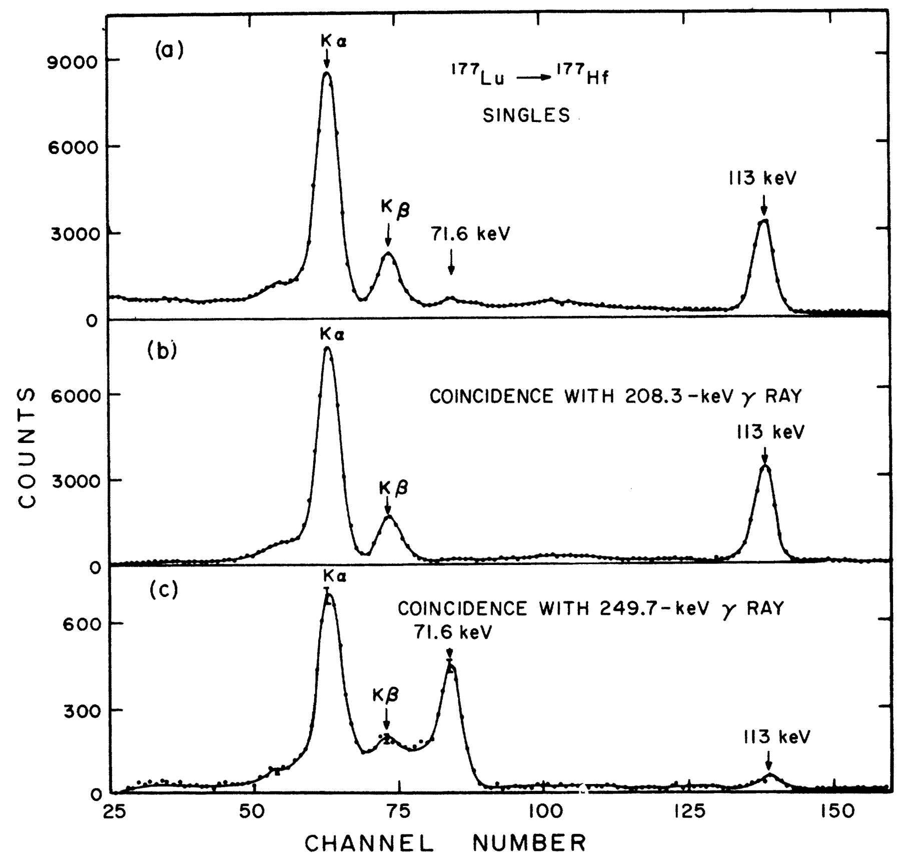

FIG. 2. The low-energy spectrum of ^{177}Lu in a 1-cm^3 Ge(Li) detector with a thin window—(a) singles spectrum, (b) spectrum in coincidence with the 208.3-keV γ ray, and (c) coincidence with the 249.7-keV γ ray (see text).

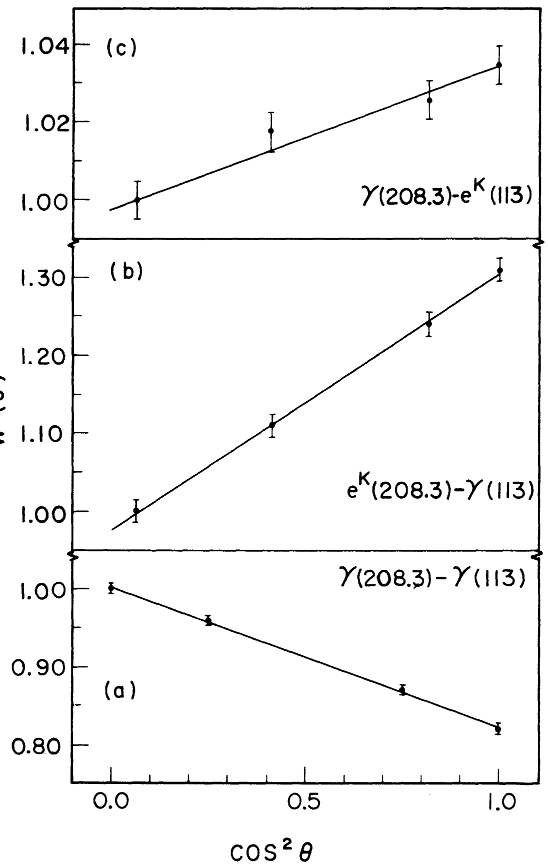

FIG. 3. The observed angular correlations of the 208.3-113-keV cascade— (a) $\gamma(208.3)$-$\gamma(113)$, (b) $e^K(208.3)$-$\gamma(113)$, and (c) $\gamma(208.3)$-$e^K(113)$. The solid lines are the least-squares-fit curves of the angular correlation functions.

rected for the β background are shown in Fig. 3. The angular correlation coefficients A_2 were obtained through a least-squares fit of the experimental points to the function $W(\theta) = 1 + A_2P_2(\cos\theta)$. The A_2 coefficients corrected for the finite solid angles of the detectors are listed in Table III.

F. γ-γ angular correlations

The $\gamma(208.3)$-$\gamma(113)$-keV angular correlation was measured for various source conditions using a Ge(Li)-NaI(Tl) coincidence system. A 20-cm^3 Ge(Li) detector was used as the fixed detector kept at a distance of 6 cm from the source. The movable detector was a 4.4-cm-diam × 5-cm-thick NaI(Tl) crystal mounted on an RCA 6810A photomultiplier and kept at a distance of 7 cm from the source. Lead cones were used in front of the detectors to minimize effects of scattering. Absorbers of 2-mm cadmium and thin copper foils were used in front of the detectors to reduce the intensity of x rays. A conventional fast-slow coincidence unit with a resolving time of 60 nsec was used. The coincidence spectra were recorded at angles 90, 120, 150, and 180° in four quadrants of a 512-channel analyzer. The energy gates in the NaI(Tl) detector were kept the same for both the e^--γ and the corresponding γ-γ angular correlations. The 208.3-113-keV γ-γ angular correlation was measured in two ways: (i) The 208.3-keV γ ray was detected in the Ge(Li) detector and the 113-keV γ ray was detected in the NaI(Tl) detector; (ii) the 208.3-keV γ ray was detected in the NaI(Tl) and the 113-keV γ ray in the Ge(Li) detector. The A_2 coefficients in both cases were found to be the same within experimental errors, and so the weighted mean value was taken and is shown in Table III. Solid angle corrections for the Ge(Li) detector were obtained by interpolation from the tables of Camp and Van Lehn.[27] The same source used for measuring the e^--γ angular correlation was used for measuring the γ-γ angular correlation also. The γ-γ angular correlation was also measured with the source used for e^--γ correlation, sealed in vacuum in a thin-walled Pyrex tube. No change in angular correlation due to possible change[28] in the physical form of the source in vacuum was observed. In order to get the true correlation for determining the mixing ratios, the γ-γ angular correlation was also measured with a $LuCl_3$ source dissolved in dilute HCl. The angular correlation coefficients are shown in Table III.

TABLE III. Results of angular correlation measurements of the 208.3-113-keV cascade.

Cascade	Form of source	A_2
$\gamma(208.3)$-$\gamma(113)$	Solution in dilute HCl	-0.192 ± 0.007
$\gamma(208.3)$-$\gamma(113)$	Solid in air	-0.150 ± 0.003
$\gamma(208.3)$-$\gamma(113)$	Solid in vacuum	-0.149 ± 0.007
$e^K(208.3)$-$\gamma(113)$	Solid in vacuum	$+0.231 \pm 0.007$
$\gamma(208.3)$-$e^K(113)$	Solid in vacuum	$+0.031 \pm 0.003$

III. ANALYSIS OF THE RESULTS

The results for the 71.6-, 208.3-, and 321.3-keV (predominantly $E1$) transitions were analyzed for evaluating any $M2$ admixture and possible penetration parameters, λ_1 and λ_2. The details of the analysis are given in Ref. 29. In the case of the 71.6- and 321.3-keV transitions a least-squares fit of the conversion coefficients were made. For the 208.3- and 113-keV transitions the conversion coefficients and the particle parameters b_2^K were analyzed by the least-squares method. The conversion coefficient of an $E1+M2$ transition in the ith shell can be expressed as[30]

$$\alpha^i(\text{Expt}) = Q\beta^i(M2) + (1-Q)\alpha^i(E1)[1 + A_1^i(E1)\lambda_1 + A_2^i(E1)\lambda_1^2 + A_3^i(E1)\lambda_2 + A_4^i(E1)\lambda_2^2 + A_5^i(E1)\lambda_1\lambda_2], \quad (2)$$

where $Q=\delta^2/(1+\delta^2)$ is the $M2$ admixture, δ is the multipole mixing amplitude, and $\alpha^i(E1)$ and $\beta^i(M2)$ are the theoretical[21] $E1$ and $M2$ conversion coefficients without penetration effects. The $A_n^i(E1)$ coefficients are tabulated in Ref. 30. λ_1 and λ_2 are the penetration parameters[30] containing the $\int \vec{j}_n \cdot \vec{r}$ and $\int \vec{j}_n \cdot \vec{\nabla}$ type penetration matrix elements. The $\int \vec{j}_n \cdot \vec{\nabla}$ type matrix element is forbidden in the asymptotic quantum numbers[5,6] for these three $E1+M2$ transitions so that the values of λ_2 are expected to be small for these transitions.

The angular correlation between the $e^K(208.3)$-$\gamma(113)$ cascade [neglecting terms involving $P_4(\cos\theta)$ and higher order] can be written[21,30] as

$$\begin{aligned} W(\theta, e_1^K-\gamma_2) &= 1 + G_2' A_2(e_1^K-\gamma_2)P_2(\cos\theta) \\ &= 1 + G_2' A_2(e_1^K)A_2(\gamma_2)P_2(\cos\theta). \end{aligned} \quad (3)$$

We define the particle parameter for the 208.3-keV transition as

$$b_2^K(208.3) = \frac{A_2(e^K 208.3)}{A_2(\gamma 208.3)} = \frac{A_2(e_1^K-\gamma_2)}{A_2(\gamma_1-\gamma_2)}. \quad (4)$$

The parameters $b_2^K(L, L')$ and the coefficients $A_n(E1)$, $C_n(E1)$, $E_n(E1)$, $B_n(M1)$, $D_n(M1)$, and $F_n(M1)$ are obtained by interpolation from the tables of Hager and Seltzer.[21,30] The $F_2(LL'II')$ coefficients are given by Frauenfelder and Steffen.[31] We define the particle parameter for the 113-keV transition as

$$b_2^K(113) = \frac{A_2(e^K 113)}{A_2(\gamma 113)} = \frac{A_2(\gamma_1-e_2^K)}{A_2(\gamma_1-\gamma_2)}. \quad (5)$$

If the attenuation coefficients G_2' and G_2 are the same in the e^K-γ and γ-γ angular correlations, the particle parameter defined as above is the same as the ratio of the experimental directional correlation coefficients $A_2(e^K$-$\gamma)$ and $A_2(\gamma$-$\gamma)$. In Sec. II F we have shown that there is no effect of vacuum on the attenuation coefficient. Also the after effects of internal conversion are expected to be negligible as the source was deposited on a conducting backing.[32] Furthermore, since the same source was used both for e^--γ and γ-γ angular correlations, it is reasonable to assume that $G_2 \approx G_2'$. Therefore, the particle parameter b_2^K can be obtained as the ratio of the experimentally measured e^K-γ and γ-γ directional correlation coefficients. The results obtained for $b_2^K(113)$ and $b_2^K(208.3)$ are:

$$b_2^K(113) = -0.20 \pm 0.02$$

and

$$b_2^K(208.3) = -1.54 \pm 0.06.$$

The measured conversion coefficient for the 113-keV transition including penetration effects can be expressed by the equation

$$\begin{aligned} \alpha^i(\text{Expt}) = Q\alpha^i(E2) + (1-Q)\beta^i(M1) \\ \times [1 + B_1^i(M1)\lambda + B_2^i(M1)\lambda^2]. \end{aligned} \quad (6)$$

The conversion coefficients $\alpha^i(E2)$ and $\beta^i(M1)$, without penetration effects, are obtained by interpolation from the tables of Hager and Seltzer.[21]

A. 71.6-keV transition

The measured values of the conversion coefficients for the 71.6-keV transition are listed in Table II. The values of Q, λ_1, and λ_2 were obtained by a least-squares fit of the measured quantities α^K, $\alpha^{L\text{I}}$, $\alpha^{L\text{II}}$, and $\alpha^{L\text{III}}$. The only physically significant set of solutions for Q, λ_1, and λ_2 is shown as (a) in Table IV. The set of solutions (a) is consistent with the prediction that λ_2 should be small.

The mixing ratio of the 71.6-keV transition has been determined to be $\delta = -0.0173 \pm 0.0065$, corresponding to $Q = 0.0003 \pm 0.0002$ by Hrastnik *et al.*[16] from the measurement of 71.6-249.7-keV γ-γ angular correlation using two Ge(Li) detectors. The present value of Q is in agreement with this. Taking the more accurate value of Q from angular correlation measurement, a least-squares fit for λ_1 was made keeping $\lambda_2 = 0$. This is shown as solution (b) in Table IV. This set of solutions has been used for calculating the matrix elements in Sec. IV.

B. 321.3-keV transition

The K-shell and L-subshell conversion coefficients for the 321.3-keV transition measured in the present work are listed in Table II. The present value of α_K is in fair agreement with that of Hager and Seltzer,[9] but is in disagreement with the value obtained by Bashandy, El-Sayad, and El-

TABLE IV. Results of least-squares fit for the 71.6-keV transition.

	Q(%)	λ_1	λ_2	χ^2
(a)	0.02 ± 0.16	3.3 ± 2.3	583 ± 657	2.63
(b)	0.03 ± 0.02	1.3 ± 0.8	···	0.88

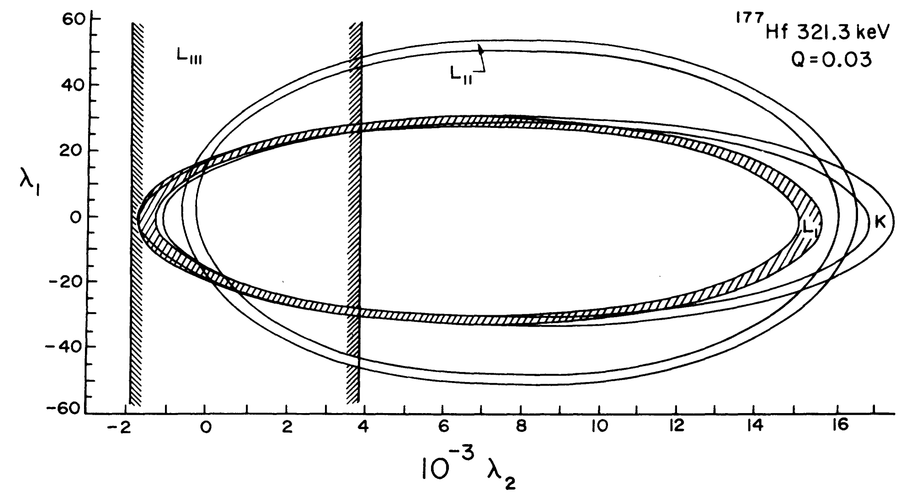

FIG. 4. Plots of the λ_1 vs λ_2 for the 321.3-keV transition for an assumed value of the $M2$ admixture, $Q = 0.03$, consistent with each of the measured conversion coefficients in the K shell and the L_{I}, L_{II}, and L_{III} subshells. Two possible areas of agreement giving two sets of solutions for λ_1 and λ_2 are indicated by the plots. The final solutions for Q, λ_1, and λ_2 are obtained by least-squares adjustment in these two regions giving two sets of final solutions.

Aassar.[8] The parameters Q, λ_1, and λ_2 were obtained through a least-squares fit of the measured quantities to Eq. (2). The initial guess values for these parameters were obtained from plots of the measured conversion coefficients in the λ_1-λ_2 plane for an assumed value of Q. A typical set for $Q = 0.03$ is shown in Fig. 4. The solutions obtained by least-squares fit are shown in Table V. The solutions (a) and (b) are those obtained for Q, λ_1, and λ_2. The solution (a) is consistent with $\lambda_2 \approx 0$ as expected from selection rules,[5,6] while (b) gives nonzero λ_2. If λ_2 is now kept fixed as zero, the least-squares-fit solutions for Q and λ_1 are given in (c) and (d). Solution (c) is equivalent to (a). The present value of λ_1 is in agreement with the earlier analysis of Pauli and Alder.[33]

C. 208.3- and 113-keV transitions

The measured values of the K-shell and L-subshell conversion coefficients of the 208.3- and 113-keV transitions are listed in Table II. The values of the particle parameters are given in Sec. III. The experimental values of the internal conversion coefficients and particle parameter for the 113-keV transition reported by Holmberg *et al.*[14] are in agreement with the present measurements. The 208.3-113-keV γ-γ angular correlation coefficient using a $LuCl_3$ source in solution in the present work (Table III) is in agreement with the value reported by Hrastnik *et al.*[16] using two high resolution Ge(Li) detectors, but is in disagreement with the value reported by Holmberg *et al.*[14] There is considerable attenuation of the correlation when a solid source is used. The γ-γ angular correlation with a $LuCl_3$ source in solution can have attenuation due to a time-dependent quadrupole interaction. However, due to the short lifetime ($T_{1/2} = 0.5$ nsec) of the 113-keV state, the attenuation of the angular correlation is expected to be very small (i.e., $G_2 > 0.99$) even for a fairly strong quadrupole interaction.[31,34] This is in agreement with the limit of the attenuation factor experimentally obtained using sources of different viscosities.[19]

Figure 5 shows plots of the measured conversion coefficients and particle parameter for the 208.3-keV transition in the λ_1-δ plane keeping $\lambda_2 = 0$. Possible solutions for δ and λ_1 were indicated for the sets of values $\delta(208.3) \approx -0.04$, $\lambda_1 \approx -1.5$ and $\delta(208.3) \approx +0.04$, $\lambda_1 \approx 0$. Similar plots for the 113-keV transition in the λ-δ plane (not shown) indicated solutions around $\lambda \approx +5$ and $\delta(113) \approx -4.0$. Least-squares fit of the conversion coefficients, the particle parameters of the 208.3- and 113-keV transitions, and the γ-γ angular correlation coefficient A_2 was made to extract the $E1$ penetration parameters λ_1 and λ_2 of the 208.3-keV transition,

TABLE V. Results of the least-squares fit for the 321.3-keV transition.

	Q(%)	λ_1	λ_2	χ^2
(a)	3.1 ± 1.6	-19.2 ± 1.2	250 ± 240	0.096
(b)	3.6 ± 1.8	15.8 ± 1.2	776 ± 234	0.16
(c)	3.6 ± 1.5	-18.2 ± 0.8	0	0.099
(d)	1.8 ± 5.1	17.3 ± 2.4	0	0.95

the $M1$ penetration parameter λ for the 113-keV transition, and the amplitude mixing ratios $\delta(208.3)$ and $\delta(113)$ by using Eqs. (2)-(6). Initial guess values of these parameters were obtained from the plots as mentioned above. A unique set of solutions was obtained from the least-squares fit. This is shown as (a) in Table VI. This showed a large value of λ_2 contrary to what is expected from selection rules. When λ_2 was kept fixed as a zero, the least-squares fit gave the set of solutions (b). Except λ_2 all the other parameters in set (b) are in agreement with those in set (a). The values of $\delta(113) = -4.0 \pm 0.6$ and $\lambda(113) = 9 \pm 8$ deduced from the conversion measurements of Holmberg *et al.*[14] are in agreement with the present values (see Table VI). However, when their γ-γ angular correlation measurement was also included in the analysis their final results became $\delta(113) = -4.75 \pm 0.07$ and $\lambda(113) = -0.6 \pm 2.0$. These are in disagreement with the present values. This disagreement is due to the smaller value of the magnitude of the A_2(208.3-113) γ-γ angular correlation coefficient obtained in Ref. 14 as compared to the present value and that of Hrastnik *et al.*[16] The present values of the mixing ratios and the penetration parameters can be expected to be more reliable as these are obtained from least-squares analysis of all the measured quantities.

D. 136.7-keV transition

The K-shell and L-subshell conversion coefficients of the 136.7-keV transition measured in the present work are shown in Table II. Because of the low intensity of this transition the measured conversion coefficients are not precise enough to deduce the penetration parameter and the mixing ratio simultaneously. The mixing ratio deduced from the conversion coefficients is $(95 \pm 5)\%$ $E2$ $+(5 \pm 5)\%$ $M1$, assuming $\lambda = 0$.

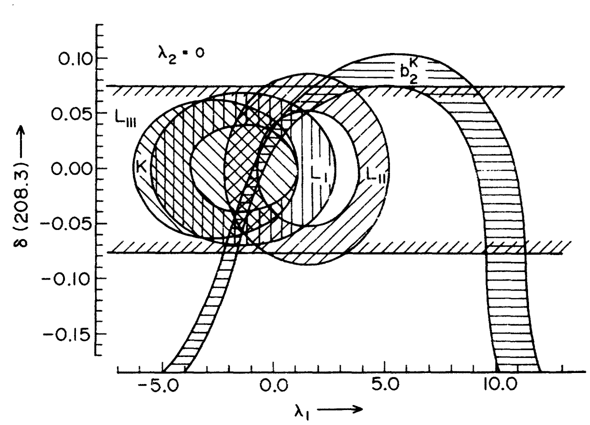

FIG. 5. Plots of the measured b_2^K, α^K, $\alpha^{L_{\rm I}}$, $\alpha^{L_{\rm II}}$, and $\alpha^{L_{\rm III}}$ for the 208.3-keV transition in the λ_1-δ plane keeping $\lambda_2 = 0$. From these plots two possible sets of solutions with $\delta \approx -0.04$, $\lambda_1 \approx -1.5$, and $\delta \approx +0.04$, $\lambda_1 \approx 0$, are indicated. The final solutions are obtained by least-squares fit using each of the above sets of solutions as initial values.

IV. DISCUSSION

It is clear from the present work that the mixing ratios of the 208.3- and 113-keV transitions can be deduced from internal-conversion measurements only after taking account of penetration effects. The mixing ratios and the penetration parameters obtained in the present work are more reliable and unambiguous as these are deduced from least-squares fit of several measured quantities.

The 321.3-keV transition, which has the largest retardation (6×10^6) of all the transitions studied in the present work, has large penetration effect in its internal conversion. The $E1$ γ-ray matrix elements and the $\int \vec{j}_n \cdot \vec{\nabla}$ type penetration matrix elements are forbidden in the asymptotic limit for the 321.3-, 208.3-, and 71.6-keV transitions, and are sensitive to effects of pairing and Coriolis coupling.[35,36] The results of the present measurements on the three transitions are consistent with $\lambda_2 \approx 0$. The $\int \vec{j}_n \cdot \vec{r}$ type penetration matrix elements and the $M2$ γ-ray matrix elements are allowed in the asymptotic quantum numbers and are expected to follow the theoretical branching rules.[37] This is found to hold good within experimental error limits. Similar results were obtained[9,38] in the case of transitions in ^{175}Lu.

From the measured half-life[39,40] of the 321.3-keV level ($T_{1/2} = 0.67$ nsec), the relative intensities of the γ rays, and the internal-conversion coefficients, the γ-ray transition probabilities could be calculated. One can then obtain the experimental $E1$ and $M2$ γ-ray matrix elements, G_{E1} and G_{M2}, from the transition probabilities using the relations

$$T_\gamma(E1, I'K' \to IK) = 3.83 \times 10^{14} (Z/A)^2 A^{1/3} E_\gamma{}^3 \times |\langle I\,1KK' - K | I\,1I'K'\rangle|^2 G_{E1}{}^2 \qquad (7)$$

TABLE VI. Results of least-squares fit for the 208.3- and 113-keV transitions.

	(a)	(b)
δ(208.3)	-0.001 ± 0.018	-0.007 ± 0.038
λ_1(208.3)	-0.68 ± 0.41	-0.82 ± 0.86
λ_2(208.3)	-168 ± 49	0
δ(113)	-4.03 ± 0.13	-3.99 ± 0.25
λ(113)	5.7 ± 5.0	5.6 ± 8.4
χ^2	0.34	0.83

TABLE VII. Propertiesof the $\frac{9}{2}^{+}[624] \to \frac{7}{2}^{-}[514]$ interband transitions in ^{177}Hf.

Quantity	321.3 keV [a]	208.3 keV [b]	71.6 keV [c]
$H_W(E1)$ [d]	6×10^6	3×10^4	9×10^4
$Q(\%M2)$	3.1 ± 1.6	≤ 0.2	0.03 ± 0.02 [e]
$\|G_{E1}\|$ (Expt)	1.4×10^{-3}	1.3×10^{-1}	2.4×10^{-1}
$H_N(E1)$ [f]	420	0.5	0.7
$\|G_{M2}\|$ (Expt)	6.8 ± 1.8	<40	49 ± 20
$H_N(M2)$ [f]	11 ± 6	>0.2	$0.2^{+0.2}_{-0.1}$
λ_1(Expt)	-19.2 ± 1.2	-0.8 ± 0.9	$+1.3\pm 0.8$ [g]
λ_2(Expt)	250 ± 240	...	...
$\|\lambda_1\|$ (Calc)	21.1	1.17	0.57
$\|\lambda_2\|$ (Calc)	44	2.5	1.19
$\|\mathfrak{M}_{pen}(\int \vec{j}_n \cdot \vec{r})\|$ (Expt)	1.7 ± 0.1	6 ± 7	20 ± 12
$\|\mathfrak{M}_{pen}(\int \vec{j}_n \cdot \vec{\nabla})\|$ (Expt)	0.3 ± 0.3	...	...

[a] From the set of solutions (a) in Table V.
[b] From the set of solutions (b) in Table VI.
[c] From the set of solutions (b) in Table IV.
[d] Hindrance factor with respect to the single-particle estimate (S. A. Moszkowski, see Ref. 31, p. 863).
[e] From Ref. 16.
[f] Hindrance factor with respect to Nilsson estimate (Ref. 41).
[g] Calculated from experiments assuming $\lambda_2 = 0$, and taking the value of Q from Ref. 16.

and

$$T_\gamma(M2, I'K' \to IK) = 2.73\times 10^6 A^{1/3} E_\gamma^{\,5} \times |\langle I2KK' - K|I2I'K'\rangle|^2 G_{M2}^{\,2}, \tag{8}$$

where the symbols have their usual meanings.[41]

E_γ is the transition energy in MeV. The experimental values of $|G_{E1}|$ and $|G_{M2}|$ for the three $E1+M2$ transitions are given in Table VII. The $E1$ γ-ray matrix element and the $\int \vec{j}_n \cdot \vec{r}$ and $\int \vec{j}_n \cdot \vec{\nabla}$ type penetration matrix elements are related to the penetration parameters λ_1 and λ_2 defined by Hager and Seltzer[30] by the relations

$$\lambda_1 \approx \bar{\lambda}_1 = \frac{\mathfrak{M}_{pen}(\int \vec{j}_n \cdot \vec{r})}{2MRG_{E1}} \tag{9}$$

and

$$\lambda_2 \approx \bar{\lambda}_2 = \frac{\mathfrak{M}_{pen}(\int \vec{j}_n \cdot \vec{\nabla})}{G_{E1}}. \tag{10}$$

The experimental values of the penetration matrix elements are given in Table VII. Theoretical values of the penetration matrix elements have been estimated using the formulation of Voikhanskii *et al.*[42] based on Nilsson-model wave functions.[41] From these and the experimental values of the γ-ray matrix elements the theoretical estimates of the penetration parameters have been calculated. The relations between the different definitions of the penetration parameters have been given by Grigorev and Feresin.[43] These have been used in the calculations of λ_1(calc) and λ_2(calc) given in Table VII. The experimental values of the matrix elements are in fair agreement with the theoretical values.

The value of the penetration parameter λ for the 113-keV transition has been estimated using the formulation of Reiner,[7] assuming that one orbital is dominant in the Nilsson wave function. The value calculated for a deformation $\eta = 4.3$ and $g_K = 0.211\pm 0.013$, $g_R = 0.253\pm 0.013$, and $g_s^{\text{eff}} = 0.6 g_s^{\text{free}}$ is $\lambda = -2.5$. This may be compared with the present experimental value of $\lambda = 5.6\pm 8.4$ given in Table VI.

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[1]P. Marmier and F. Boehm, Phys. Rev. 97, 103 (1955).
[2]P. Alexander, F. Boehm, and E. Kankeleit, Phys. Rev. 133, B284 (1964).
[3]A. J. Haverfield, F. M. Bernthal, and J. M. Hollander, Nucl. Phys. A94, 337 (1967).
[4]C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (Wiley, New York, 1967), 6th ed.
[5]S. G. Nilsson and J. O. Rasmussen, Nucl. Phys. 5, 617 (1958).
[6]E. L. Church and J. Weneser, Nucl. Phys. 28, 602 (1961); Annu. Rev. Nucl. Sci. 10, 193 (1960).
[7]A. S. Reiner, Nucl. Phys. 5, 544 (1958).
[8]E. Bashandy, M. G. Mousa El-Sayad, and M. Riad El-Aassar, Z. Phys. 186, 108 (1965).
[9]R. Hager and E. Seltzer, in *Internal Conversion Processes,* edited by J. H. Hamilton (Academic, New York, 1966), p. 315.
[10]T. Novakov and J. M. Hollander, Nucl. Phys. 60, 593 (1964).
[11]S. Hogberg, R. Jadrny, S. E. Karlsson, G. Malmsten, and O. Nilsson, Z. Phys. 254, 89 (1972).
[12]J. E. Thun, Z. Grabowski, W. D. Hamilton, and M. S. El-Nesr, Nucl. Phys. 29, 13 (1962).
[13]L. Holmberg, V. Stefansson, and M. Gunnerhed, in *Radioactivity in Nuclear Spectroscopy,* edited by J. H. Hamilton and J. C. Manthuruthil (Gordon and Breach, New York, 1972), Vol. 2, p. 1407; Phys. Scr. 4, 41 (1971); L. Holmberg and V. Stefansson, in *Angular*

Correlation in Nuclear Disintegration, edited by H. Van Krugten and B. Van Nooijen (Rotterdam U. P., 1971), p. 168.

[14]L. Holmberg, V. Stefansson, J. Becker, C. Bargholtz, and L. Gidefeldt, Phys. Scr. 6, 177 (1972).

[15]S. Tornkvist, S. Strom, J. E. Thun, V. Schmidt, and N. Baade, Nucl. Phys. A117, 336 (1968).

[16]B. Hrastnik, I. Basar, M. Diksic, K. Ilakovac, V. Kos, and A. Ljubicic, Z. Phys. 239, 25 (1970); also in *Angular Correlations in Nuclear Disintegration* (see Ref. 13), p. 251.

[17]E. D. Klema, Phys. Rev. 109, 1652 (1958).

[18]H. J. Behrend, Z. Naturforsch 13a, 211 (1958).

[19]S. Ofer, Nucl. Phys. 3, 479 (1957).

[20]H. I. West, Jr., L. G. Mann, and R. J. Nagle, Phys. Rev. 124, 527 (1961).

[21]R. S. Hager and E. C. Seltzer, Nucl. Data A4, 1, 397 (1968).

[22]A. P. Agnihotry and K. P. Gopinathan, Nuclear Physics and Solid State Physics (India), Vol. 14B, p. 431; *ibid.*, Vol. 15B, p. 393.

[23]V. N. Grigorev, D. M. Kaminkar, and Yu. V. Sergenkov, Izv. Akad. Nauk SSSR Ser. Fiz. 36, 842 (1972).

[24]A. P. Agnihotry, K. P. Gopinathan, H. C. Jain, and C. V. K. Baba, Phys. Rev. C 6, 321 (1972).

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

[26]Y. K. Agarwal, C. V. K. Baba, and N. C. Mukhopadhyay, Nucl. Phys. A127, 661 (1969).

[27]D. C. Camp and A. L. Van Lehn, Nucl. Instrum. Methods 76, 192 (1969); 87, 147 (1970).

[28]E. W. Koopman and H. Van Krugten, Physica (Utr.) 42, 262 (1969); J. H. Trueblood and F. T. Avignone, III, Phys. Rev. C 4, 1490 (1971); W. L. Croft, D. Krumpotic, and J. H. Hamilton, in *Radioactivity in Nuclear Spectroscopy* (see Ref. 13), Vol. 2, p. 1445.

[29]A. P. Agnihotry and K. P. Gopinathan, Phys. Rev. C 6, 1394 (1972).

[30]R. S. Hager and E. C. Seltzer, Nucl. Data A6, 1 (1969).

[31]H. Frauenfelder and R. M. Steffen, In *Alpha-, Beta-, and Gamma-Ray Spectroscopy,* edited by K. Siegbahn (North-Holland, Amsterdam, 1966), p. 997.

[32]U. Baverstam, A. Johansson, and T. R. Gerholm, Ark. Fys. 35, 451 (1967).

[33]H. C. Pauli and K. Alder, Z. Phys. 202, 255 (1967).

[34]J. Berthier, P. Boyer, and J. I. Vargas, in *Hyperfine Interactions in Excited Nuclei,* edited by G. Goldring and R. Kalish (Gordon and Breach, New York, 1971), Vol. 2, p. 439.

[35]M. N. Vergnes and J. O. Rasmussen, Nucl. Phys. 62, 233 (1965).

[36]F. M. Bernthal and J. O. Rasmussen, Nucl. Phys. A101, 513 (1967).

[37]G. Alaga, K. Alder, A. Bohr, and B. R. Mottelson, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 29, No. 9 (1955).

[38]G. T. Emery and M. L. Perlman, Phys. Rev. 151, 984 (1966).

[39]E. E. Berlovich, Yu. K. Gusev, V. V. Ilin, and M. K. Nikitin, Zh. Eksp. Teor. Fiz. 43, 1625 (1962) [transl.: Sov. Phys.—JETP 16, 1144 (1963)].

[40]K. P. Gopinathan, A. P. Agnihotry, S. B. Patel, and M. S. Bidarkundi, Contributions to International Conference on Nuclear Moments and Nuclear Structure, Osaka, Japan, 1972 (unpublished), p. 237; J. Phys. Soc. Jap. Suppl. 34, 430 (1973).

[41]S. G. Nilsson, K. Dan. Vidensk, Selsk. Mat.-Fys. Medd. 29, No. 16 (1965); B. R. Mottelson and S. G. Nilsson, K. Dan. Vidensk. Selsk. Mat.-Fys. Skr. 1, No. 8 (1959).

[42]M. E. Voikhanskii, V. N. Grigorev, M. A. Listengarten, and A. P. Feresin, Izv. Akad. Nauk SSSR Ser. Fiz. 35, 847 (1971) [transl.: Bull. Acad. Sci. USSR Phys. Ser. 35, 780 (1971)].

[43]V. N. Grigorev and A. P. Feresin, Yad. Fiz. 12, 665 (1970) [transl.: Sov. J. Nucl. Phys. 12, 361 (1971)].