Electromagnetic Transitions in $^{177}\text{Hf}$

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The gamma transitions in $^{177}\text{Hf}$ have been measured using a 50 cm radius iron-free double focusing beta-ray spectrometer. The decay scheme is investigated and the K-conversion coefficients of the 208 and 321 keV transitions have been determined.

Comparison with theoretical conversion coefficients for pure E1 radiation calculated by Sliv and Band indicates that the K-conversion process of the 208 keV transition is normal while the large anomaly observed in the K-conversion process of the 321 keV E1 transition is compatible with a large M2 admixture or more probable with the presence of penetration matrix elements. From comparison with theoretical conversion coefficients the 113 keV M1 + E2 transition has been found to be (95.12 ± 0.6)% E2 corresponding to $\mid \beta \mid = 4.38 ± 0.33$. The decay scheme of $^{177}\text{Lu}$ is discussed in terms of nuclear models.

Introduction

The unified model has been used with reasonable success in interpreting the properties of the lower levels of $^{177}\text{Hf}$. According to the formulation of $^{177}\text{Hf}$, it should exhibit a well developed rotational structure. It is, therefore, of interest to apply nuclear spectroscopy techniques to investigate the characteristics of the $^{177}\text{Hf}$ nucleus. The strongly deformed odd mass nuclei are of great interest from the theoretical point of view. Bohr and Mottelson have proved that the unified model is very successful in explaining nuclear levels and transition probabilities in strongly deformed nuclei. One observes in strongly deformed odd mass nuclei both particle excited states and associated rotational excitations. The odd mass rotational states depend more specifically on the properties of the ground state, they do not exhibit the same simple trends as those in even nuclei. Mottelson and Nilsson have performed analysis of the intrinsic states of odd mass nuclei having an ellipsoidal equilibrium shape. They have described the relevant levels in terms of the asymptotic quantum notation and have classified experimentally observed levels.

The spin of the ground state of $^{177}\text{Hf}$, see Figure 1, was measured by Speck and Jenkins to be 7/2 which is in agreement with the strong coupling estimate of Mottelson and Nilsson. Rotational levels associated with this intrinsic configuration have been observed at 113 keV ($I = 9/2$) and 250 keV ($I = 11/2$). An intrinsic excitation has been observed at 321 keV in $^{177}\text{Hf}$, its spin has been determined from angular correlation as 9/2. It decays by E1 transitions to the members of the ground state rotational band and the configuration is clearly identified as 9/2. In the Nilsson asymptotic description the configuration of the 321 keV level is 9/2 + [624]. The 113 keV (M1 + E2) transition in $^{177}\text{Hf}$ has been studied by many authors. The M1 component is strongly retarded due to cancellation between the g-factors for particle structure and rotation, and penetration may thus affect the internal conversion of the transition.

From the K and L conversion coefficients and from the angular correlation measurement it was shown that the 208 keV transition is mostly E1 with no more than 2% admixture of M2. The cross over of 250 keV is pure E2 transition. The K-conversion coefficient of the 321 keV transition was determined and the result obtained is in disagreement with other authors.

Fig. 1. The decay scheme of $^{177}\text{Lu}$. 

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with the theoretical values for pure E1 transition calculated by Sliv and Band.  

Due to the increased interest from the theoretical point of view, to study strongly deformed odd mass nuclei, it was decided to investigate the gamma transitions in the decay of $^{177}\text{Hf}$ carefully using a high resolution 50 cm radius iron free double focusing beta-ray spectrometer.

1. Experimental Procedures

1.1. Apparatus

In the present experiments we used the iron free double focusing beta-ray spectrometer described elsewhere (R0 = 50 cm), to study the electron spectrum of gamma transitions in $^{177}\text{Hf}$. The detector employed is a G.M. counter with 1.8 mg/cm$^2$ mica end-window.

1.2. Source Preparation

The sources were produced by neutron irradiation of natural spectroscopically pure lutethium oxide and lutethium chloride in the E.A.R. reactor at Inchass in a flux of about 10$^{13}$ neutrons cm$^{-2}$ sec$^{-1}$.

For the internal conversion source, the Lu$_2$O$_3$ was transferred into LuCl$_3$ and evaporated in vacuum onto an aluminium foil of thickness about 1 mg/cm$^2$. The evaporated material was uniformly distributed in a rectangular form of dimensions $0.15 \times 1.5$ cm$^2$. The thickness of the material deposited was about 100 µg/cm$^2$.

As external conversion source, Lu$_2$O$_3$ was enclosed in a spectroscopically pure aluminium capsule. The inner dimensions of the capsule were 1 mm diameter and 15 mm length. The wall thickness was 0.5 mm. After the irradiation the aluminium capsule was enclosed in a copper tube with a wall thickness of 0.5 mm to stop the electrons emitted by the source.

A uranium converter 2.9 mg/cm$^2$ ($5 \times 30$ mm$^2$) was used. The converter foil was obtained from the same supply that was prepared for similar studies. The illustrative drawing of source holder for external conversion studies, which was used in other investigations carried out at this laboratory, is shown in Figure 2.

1.3. Internal Conversion Experiments

The energy of the 84 keV transition in $^{170}\text{Yb}$ and the energy of the 412 keV transition in $^{198}\text{Hg}$, carefully determined before, were used for calibration purposes. During the measurements, the spectrometer current was continuously checked using a precision potentiometer. The elimination of the earth magnetic field and other disturbing magnetic fields was also continuously checked and variations were compensated for. The data recorded were corrected for counter dead-time, constant counter background and decay. A correction for the momentum dependence of the spectrometer window width was performed by dividing the counting rate with the current for each measured point. The window width correction was performed by a division of each line intensity with the current for the line position. The tails of the conversion lines range several keV below the peak position even when extremely thin sources are used. The contribution to the conversion line intensity from the tail is considerable and to obtain an accurate measure of the total line intensity a precise determination of the background is necessary.

In deducing the areas of partially resolved lines we have found it convenient to subtract the background and beta continuum rates, and the line shapes determined from clearly resolved lines of comparable energy and appropriate to the conversion shells in question are fitted to the data in this form. All closely spaced lines, except K-lines, were assumed to have the same shape. This shape narrows quite gradually with increasing momentum; the contribution from energy degradation being less serious at high energies. For weaker lines the intensity was simply assumed to be proportional to the height compared with that of a neighbouring line of measured area. The presence of Auger lines on both sides of the 113 keV K line and the non-linear beta background below all conversion lines make such a precise determination very difficult. The L and M subshell intensity ratios were determined in the same way as determined by Karlsson et al. based on the assumption of equal line shapes for all lines in a subshell group. However, by using a modified version of the method applied to the subshell ratio
1.4. Photoelectron Experiments

The external conversion spectra for gamma energy greater than 200 keV were measured in order to determine the relative gamma intensity for the transitions found from the internal conversion study. The emission of photoelectrons from the converter is not isotropic, and in order to reduce scattering effects the external conversion process must then take place in very thin layers of materials. The photo intensity from a converter of uniform density is proportional to the total cross section $\tau_K$ (for the particular atomic shell and transition energy) and to a factor $f$, which depends on the character of the appropriate photoelectric angular distribution and on the details of the experimental arrangement of the gamma ray source and the photoelectric converter inside the spectrometer. Novakov et al.\textsuperscript{14} has developed the analysis, the $K/\Sigma L$, $\Sigma L/\Sigma M$ conversion intensity ratios could be determined with good accuracy. The internal conversion electron spectra of gamma transitions are shown in Figures 3, 4, 5, 6, and 7.
procedure of calculating the $f$-factors from the measured angular distributions for various source-geometries.

If a point source is used the $f$-factor is given by:

$$ f = \frac{\int_0^\pi f(\theta) \sin \theta \, d\theta}{\int_0^\pi f(\theta) \, d\theta} \left( 1 + \frac{\pi}{4} \tan \theta \right). $$

For a rectangular source and converter, which is the most useful geometry for measurements with a
magnetic spectrometer of a double focusing type, the following expression is obtained:

\[
\frac{2/\pi \int_0^\theta J(\theta - \Delta) \tan \theta \left[ \arcsin \left( \frac{\tan \theta_0}{\tan \theta} \right) - \arccos \left( \frac{\tan \theta_1}{\tan \theta} \right) \right] e^{-\mu g \cos \theta} d\theta}{\int_0^\pi J(\theta) \sin \theta d\theta}
\]

(2)

where \( J \) is the appropriate photoelectric angular distribution function. For definition of the various angular symbols the reader is referred to the article by Hultberg 18, 19.

The denominator of \( f \) is an integral over the angles used in an experiment and is a function of the given experimental set-up (i.e. source to converter distance, source and converter sizes, etc.). \( J(\theta) \) contains the angular dependence of the photoelectric process while the gamma-ray absorption is taken into account by the exponential factor. The symbol \( (\theta) \) gives the angle of incidence for the gamma ray against the converter, \( g \) represents the thickness of absorbing material between the source and the converter and \( \mu \) its absorption coefficient. \( f \) may be calculated for each energy and shell for which angular distributions are known.

The \( f \)-factors for different gamma-ray energies are calculated ** for extended plane sources and for K-shell photoconversion in this element.

The intensity of a gamma-ray can then be expressed 18, 19 as:

\[
I_\gamma = c (A_\gamma / \tau)
\]

(3)

where \( c \) is a constant depending on the converter thickness, source strength and the instrumental transmission factor and \( A_\gamma \) the measured intensity of a photoline taken as the area under the photoconversion peak after normalizing to the unit momentum interval.

The accuracy in the determination of the relative intensities of the gamma-rays depends on the energy and on the shell in which the conversion takes place. The most accurate determinations are obtained for the K-conversion of gamma-rays with an energy well above the threshold. Hence gamma intensities have been determined from the K-conversion for gamma ray above 200 keV. The accuracy of the gamma-ray intensities determined from the K-photo electrons at lower energies is impaired by the errors in the \( f \)-factors, mainly as a result of the electron scattering in the converter material. These errors were taken into consideration in the theoretical calculation of the \( f \)-factors. There is also an additional uncertainty in the gamma-ray intensities, for the low energy region, comes from the self-absorption of the gamma-rays in the source material and the corrections for this effect are very sensitive to the error in the absorption coefficient used in the calculation.

** The computation has been worked out by Dr. Hultberg as a programme on the electronic computer BESK at Stockholm. We express our sincere gratitude to him for making his programme available for us.

2. Absolute Determination of Internal Conversion Coefficients

The internal-external conversion method for the experimental determination of internal conversion coefficients is based on a straight-forward measurement of the rates of emission of internal conversion electrons and gamma-rays, belonging to the same transition. The ratio of these rates defines the internal conversion coefficient. The study of conversion electrons does not present any particular complications since the emission of conversion electrons from the source is basically isotropic. To find the gamma-ray emission rate, however, one has to resort to indirect methods such as the conversion to photoelectrons in heavy elements, e.g. U, Pb, Au, Pt and Yb. The emission of photoelectrons from the converter is not isotropic and in order to reduce scattering effects the external conversion process must then take place in very thin layers of material.

According to Ref. 20, the K-conversion coefficient can be calculated from the following formula:

\[
\alpha_K = \frac{(A_{\text{in}})_K}{(A_{\text{ex}})_K} \tau_K f_K S d b c.
\]

(4)

\( \alpha_K \) = internal K-conversion coefficient,

\( (A_{\text{in}})_K \) = recorded intensity of internal K-conversion electrons,

\( (A_{\text{ex}})_K \) = recorded intensity of K-photoelectrons,

\( \tau_K \) = tabulated value of photoelectric cross-section for K-shell (barns/atom),

\( f_K \) = the correction which accounts for the anisotropical distribution of the photoelectrons emitted from the K-shell,

\( S \) = the ratio of the external and internal sources strength,
\[ d = \text{the thickness of the converter in mg/cm}^2, \]
\[ b = \text{a dimension factor (2.531} \times 10^{-6} \text{atoms cm}^{-2} \text{barns mg for uranium)}, \]
\[ c = \text{a correction factor to account for differences in transmission of the spectrometer for the external and internal conversion sources.} \]

The basic quantity for the determination of the internal conversion coefficient is the integrated photoelectric cross section \( \tau \). It is therefore essential to have access to accurate tables of \( \tau \). At present the most accurate calculations are those for \( \tau_K \) by Grodstein \(^21\) and by Hultberg et al. \(^22\). They are corrected to any order in \( 3Z \) but neglect the effect of screening. The correction for the latter effect is rather small for the K-shell and can partially be corrected for.

The factors \( S, d, b, \) and \( c \) in Eq. 4 could be omitted by making use of the comparison method where a transition in the same isotope whose is accurately measured should compared with a transition whose \( \tau_K \) would be calculated. Because of the different line intensities and possible interference of other lines, the error in \( A_{\text{in}}/A_{\text{ex}} \) varies strongly from one transition to another.

3. Results and Discussion

The conversion spectrum of the transitions in \(^{177}\text{Hf} \) was measured several times and all lines were found to decay with 6.8 days half-life corresponding to pure \(^{177}\text{Lu} \) radioactivity. On the basis of angular correlation results \(^23\), the 113 keV radiation cannot be a pure E2 transition. If on the other hand, the 208 keV radiation is considered to be pure E1 radiation, then there is obtained as mixing parameter \( \delta \) for the 113 keV ray:

\[ -5.16 < \delta < -4.84 \]

which corresponds to an M1 admixture of 3.6-4.1%. If the 208 keV gamma ray contains a 0.5 percent M2 admixture, which seems probable according to the internal conversion measurements, a mixture of 3.4 or 5 percent M1 in the 113 keV radiation was obtained \(^23\), depending upon the sign chosen for the 208 keV mixing parameter. The latter value is in agreement with the internal conversion measurements \(^23\) and with West et al. \(^24\), who obtained \( \delta_1 = -0.07 \pm 0.03 \), and \( \delta_2 = -4.0 \pm 0.2 \).

From our measurements, the conversion line intensity ratios experimentally obtained are given in Table 1. The mixing ratio \( \delta^2 = E2/M1 \) is calculated from the \( L_{11}/L_{11} \) subshell ratio according to the formula:

\[ E2/M1 = \frac{(2L_{11})M1[(L_{11}/L_{11})_{\text{exp}} - (L_{11}/L_{11})_{\text{th}}]}{(2L_{11})E2[(L_{11}/L_{11})_{E2} - (L_{11}/L_{11})_{\text{exp}}]}. \]  

The first three lines in Table 2 give the multipole mixtures of the 113 keV transition obtained when we compared the experimental data with theoretical conversion intensity ratios. The tabulations by Sliv and Band \(^11\) and Hager and Seltzer \(^25\) and that of Pauli \(^26\) have been used. In the latter two cases the nuclear effects have been taken into consideration. In Table 2 the different ratios used give somewhat different E2 percentages. For pure E2 transitions in deformed nuclei, deviations of 5-6% between theoretical and experimental \( L_{11}/L_{11} \) ratios have been established \(^27\). The lines 4-6 in Table 2 give the E2 percentages obtained when the theoretical E2 \( L_1 \) and M1 conversion coefficients are increased by 5% which agree with Hager and Seltzer prediction \(^28\). In
earlier studies of the L subshell ratios of pure E2 transitions, the experimental $L_{11}/L_{111}$ values were consistent with the theories of Rose and Sliv and Band. If, however, the more recent ICC's of Hager and Seltzer and Pauli are used, the experimental $L_{11}/L_{111}$ ratios are found to be generally lower (of the order of 2%) than the theoretical values. The internal conversion process of the 113 keV transition might be affected by penetration effects on the M1 conversion rate. However, since the E2 component completely dominates, no statements about penetration effects can be made on the basis of the present experimental data. From the E2 percentage values deduced from the $K/E2$, $L_{11}/L_{111}$, $L_{1}/L_{111}$ ratios (lines 4 and 6 in Table 2), a mean value of (95.12 ± 0.6)% E2 is obtained for the 113 keV transition in $^{177}$Hf. This corresponds to $|\delta| = 4.38 ± 0.33$, a value which is in agreement with the result $|\delta| = 4.5 ± 0.3$ obtained by Hogberg et al. and with the result $\delta = -4.0_{-1.0}^{+1.0}$ obtained by Holmberg et al. in their $e_K - \gamma$ correlation measurements.

Simons et al. measured the 72 keV - 250 keV correlation and found that there is a mixing parameter for the 72 keV radiation $0.027 < \delta < 0.014$ which corresponds to a 0.07 percentage M2 admixture at most, in this case the 250 keV radiation is considered to be a pure E2 radiation. From the 208 keV - 113 keV correlation, Simons et al. arrived at the M2 admixture in the 72 keV radiation attaining a maximum of 0.1 percent. This in good agreement with Ofer's measurement of the K-conversion coefficient of the 72 keV gamma-ray. This measurement permits an M2 admixture up to 0.8 percent. If one takes into consideration the measurement by Marmier and Boehm of the internal conversion coefficients of the 250 keV gamma-ray, one gets 5.5 → 11.5 percent M3 admixture in this transition. To fit the experimental result of the correlation measurement Simons et al. found that the 72 keV gamma ray should contain 0.2 → 0.6 percent M2 admixture which also is in agreement with Ofer's measurement of the 72 keV gamma ray.

Our experimental values for L-subshell ratios of the 72 and 208 keV transitions in $^{177}$Hf are compiled in Table 3. In our analysis we have assumed that the $L_{111}$ conversion coefficient is not affected by the penetration effects. Experiments on low energy E1 transitions in heavy nuclei have shown that for these transitions this assumption is valid even when very anomalous values are observed for the $L_4$ and $L_{11}$ conversion coefficients. Our measurement on the 208 keV transition in $^{177}$Hf gave a value for the K-conversion coefficient equal:

$$z_K(208) = 0.0445 ± 0.0089.$$  

This is in good agreement with the theoretical value by Sliv and Band:

$$z_K(208) = 0.0450,$$

for pure E1 multipolarity and also in good agreement with the experimental values $z_K(208) = 0.044 ± 0.015$ measured by Marmier and Boehm and $z_K(208) = 0.042 ± 0.015$ measured by McGowan et al.

The retardation factor for the 208 keV transition in $^{177}$Hf is equal to 4.48 x 10^2. Since the K-conversion process of the 208 keV radiation is normal and not affected by nuclear structure effects, this tends to support Nilsson and Rasmussen's statement that the nuclear structure effects on the internal conversion process of retarded E1 transition will not show up below a certain threshold in the retardation factor. Our conclusion is supported by the angular correlation measurements carried out by Thun et al. for the 208 keV - 113 keV cascade in $^{177}$Hf. They found that the angular correlation between the K-conversion electrons of the retarded 208 keV E1 transition and the 113 keV gamma rays is normal while the conversion process of the 282 keV transition in $^{175}$Lu, which is very similar to the 208 keV transition in $^{177}$Hf but has much higher retardation factor, seems to be affected by nuclear structure effects.

Another support for our conclusion was the experiment performed by Marklund who determined the $z_K$, $K/L$ and L-subshell ratios for the transitions which de-excite the 321 keV level in $^{177}$Hf. For the

Table 3. Experimental values for L-subshell ratios of the 72 and 208 keV transitions in $^{177}$Hf.

<table>
<thead>
<tr>
<th>$E_\gamma$(keV)</th>
<th>Ratio</th>
<th>Experimental value</th>
<th>Theoretical value for E1 transitions from Sliv and Band</th>
</tr>
</thead>
<tbody>
<tr>
<td>72</td>
<td>$L_{1}/L_{111}$</td>
<td>3.1 ± 0.5</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>$L_{1}/L_{111}$</td>
<td>1.2 ± 0.3</td>
<td>0.84</td>
</tr>
<tr>
<td>208</td>
<td>$L_{1}/L_{111}$</td>
<td>6.5 ± 0.6</td>
<td>5.8</td>
</tr>
<tr>
<td></td>
<td>$L_{11}/L_{111}$</td>
<td>1.05 ± 0.1</td>
<td>0.98</td>
</tr>
</tbody>
</table>

* The $\gamma$-ray matrix elements are quoted relative to Weisskopf units.
208 keV transition the data are compatible with Sliv's values which are in agreement with our results also with the angular correlation results 38.

Our measurement on the 321 keV transition gave a value for the K-conversion coefficient:

$$a_K(321) = 0.1615 \pm 0.0163$$

which is in good agreement with Bashandy's 10 value: $$a_K(321) = 0.162 \pm 0.016$$, but in disagreement with the theoretical value for pure E1 transition calculated by Sliv and Band 11:

$$a_K(321) = 0.0154$$.

The experimental value is higher by ten times than the theoretical value. Our result is also compatible with the $$a_K(321) = 0.2$$ determined by Marmier et al. 34.

The partial mean-life of the 321 keV level for a ground state gamma transition is given 40 by:

$$\tau_s(321 \text{ keV}) = 1.7 \times 10^{-8} \text{ sec}$$,

considering the intensity ratio 34 70 : 1 for the 208 keV to 321 keV gamma decay modes. When the experimental transition probability is compared with the gamma ray transition probability in terms of the single particle model 41, it was found that the 321 keV transition is retarded by factor $$2.6 \times 10^6$$. The photoelectric cross-section $$\tau_K$$ was taken from the interpolation values tabulated by White 21. $$A_{\text{in}}/A_{\text{ex}}$$ was determined with an accuracy of 3%. The estimated uncertainty in the calculated photoelectric cross-sections tabulated by White 21 was indicated by her to be 5 – 15% without giving fuller accounts. According to Hultberg and Stockendar 20, 42 the errors in $$\tau_K$$ was taken to be 6% in the present work.

The large discrepancy of the $$a_K(321 \text{ keV})$$ result is due either to an admixture of magnetic quadrupole content, or probably to the appearance of nuclear structure effects in the internal conversion process of such a retarded E1 transition. The ground state spin has been established as $$7/2$$ from atomic spectrum measurements 3 and is in agreement with the calculations of Mottelson and Nilsson 1 which also indicate that it has negative parity. The spin and parity of the 321 keV level has been determined as $$9/2$$ from the anisotropy of the 72 keV – 250 keV gamma cascade 9, and from the electron conversion coefficients 9, 34 of the 72 keV and 208 keV transitions. The hinderance factor for the ground state transition [321 keV gamma-ray decays from $$9/2 + (624)$$ to $$7/2 - (514)$$] is greater than $$10^6$$ and is in good agreement with the values tabulated by Mottelson and Nilsson 1. It is assumed that the M2 radiation in the transition is unhindered. Then, from a comparison 40 of the E1 and M2 transition probabilities, and admixture corresponding to $$\delta^2 = 0.18$$ for the M2 : E1 intensities would be expected to be present in the 321 keV transition. According to such an admixture the K-conversion coefficient could be estimated from:

$$a_K = [\beta^2 + a_1(K)]/[1 + \delta^2]$$ (6)

where $$\beta^2(K)$$ is the theoretical K-conversion coefficient for a pure M2 transition while $$a_1(K)$$ is the theoretical K-conversion coefficient for a pure E1 transition. The result obtained according to the admixture assumption is $$a_K(321) = 0.0866$$ which is still in disagreement with our experimental result:

$$a_K(321) = 0.1615 \pm 0.0163$$.

Hence the large anomaly found in the K-conversion coefficient of the 321 keV retarded E1 transition (about 10 times higher than the theoretical value) could be attributed to nuclear structure effects. Our conclusion is supported by the results obtained by Marklund 39 who showed that the conversion processes of the 72 keV and 321 keV transitions deviate from that expected theoretically. The 72 keV L-subshell ratios can be explained as arising from an anomalous conversion process with increased $$L_1$$ and $$L_{11}$$ conversion coefficients, while the observed high $$a_K(321)$$ value is compatible with a large M2 admixture or, more probably, with the presence of penetration matrix elements. One should notice that the retardation factors of the 72 keV and 321 keV transitions are at least one order of magnitude higher than for the 208 keV transition.

Our measurements confirm the level scheme of 177Hf presented in Figure 1, the spins and the parities stated there. The three lowest levels form a rotational band according to the Bohr-Mottelson unified model.

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