

continuing interest and direction during these studies. Thanks are due to L. O. Love, W. K. Prater, F. M. Scheitlin, and W. A. Bell of the Oak Ridge National Laboratories (ORNL) who did the isotopic separation and recovery of the sample, and to W. D. Harmon of

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## Directional Correlation Measurements in $\text{Hf}^{178}$

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Measurements of the directional correlations of the 1335-93 kev, 1345-93 kev, and 1390-93 kev  $\gamma$ - $\gamma$  cascades in  $\text{Hf}^{178}$  have been made, using a liquid source of 21-day  $\text{W}^{178}$  in equilibrium with 9.3-min  $\text{Ta}^{178}$ . It is shown that the data support the tentative spin assignments of Gallagher *et al.* The 1390-kev  $\gamma$  ray is found to be almost pure quadrupole radiation. If the spin and parity of the 1430-kev level is  $1+$ , as suggested by Gallagher *et al.*, the multipolarity of the 1335-kev  $\gamma$  ray is determined by our measurements to be mainly  $M1$ . Using these results, the amount of  $E0$  radiation in the 1309-kev transition has been estimated. It is found that approximately 70% of the 1390-kev  $K$ -conversion electrons are due to monopole transitions.

THE decay of the 9.3-min  $\text{Ta}^{178}$  isomer has recently been investigated by Gallagher, Nielsen, and Nielsen<sup>1</sup> (in the following referred to as GNN). Their

results indicate that about 95% of the decays proceed by direct transitions to the ground state and to the first-excited state of  $\text{Hf}^{178}$ , the remaining decays populating high-lying levels of this nucleus in the energy region 1200-1500 kev. Analyzing their internal conversion data, GNN were able to show rather unambiguously that two of these high-lying levels have spin and parity  $0+$ , but their conclusions regarding the properties of the other levels in this group were less precise, mainly due to insufficient  $\gamma$ -ray resolution. The level scheme proposed by GNN is shown in Fig. 1, and in the following we shall assume that this correctly represents the main features of the decay.

The purpose of this investigation is to obtain additional evidence on the spins of some of the high-lying levels of  $\text{Hf}^{178}$  by means of  $\gamma$ - $\gamma$  directional correlation techniques and at the same time to collect as much information as possible about the multiplicities of the corresponding decay radiations. It is apparent from Fig. 1 that a number of  $\gamma$ - $\gamma$  cascades are available in  $\text{Hf}^{178}$ , all proceeding through either the first or the second rotational state. In this experiment we have, however, confined ourselves only to the cascades involving the 93-kev photon and the  $\gamma$  rays in the region 1335-1390 kev, since it was found that the other cascades were either too weak to give a measurable correlation or too much affected by background radiations to yield valuable information.

The source consisted of about  $4 \mu\text{C}$  of 21-day  $\text{W}^{178}$  in equilibrium with 9.3-min  $\text{Ta}^{178}$ , produced in the same way as described in GNN. In order to minimize extranuclear attenuation effects, the tungsten was dissolved in hydrofluoric acid, thus forming  $\text{WF}_6$  ions. The measurements were carried out using  $1\frac{1}{2} \times 1\frac{1}{2}$  in. NaI

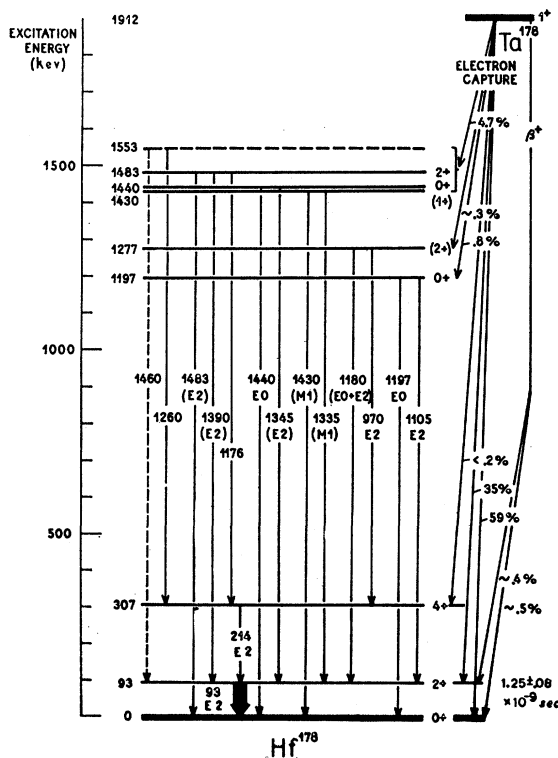


Fig. 1. Decay scheme of 9.3-min  $\text{Ta}^{178}$  according to GNN.

<sup>1</sup> C. J. Gallagher, H. L. Nielsen, and O. B. Nielsen, Phys. Rev. 122, 1590 (1961).

crystals, a fast-slow coincidence circuit with a resolving time of  $3 \times 10^{-8}$  sec, and a 256-channel analyzer. The output pulses from one of the slow linear amplifiers were fed into the analyzer which was gated by the coincidence signals. The multichannel analyzer was included in the circuit for the following reason. The  $\gamma$ -ray spectrum in coincidence with the 93-keV photon contains a broad peak with an average energy of about 1360 keV and it is known from GNN that this peak is associated with three transitions of energies 1335, 1345, and 1390 keV (see Fig. 1). The two lowest transitions appear totally unresolved in the NaI detector, but the 1390-keV photon is sufficiently separated from the other two to allow an approximate determination of its correlation function by an analysis of the upper part of the composite peak. Therefore, the multichannel analyzer was used to record pulses corresponding to high energy events above 600 keV in one counter, whereas the other counter was coupled to a single-channel analyzer set to accept events between 80 and 100 keV.

The data were recorded at the  $90^\circ$ ,  $135^\circ$ , and  $180^\circ$  positions of the movable counter, and a total of about 5000 coincidences was recorded at each angle. The real coincidence rate was normalized to correct for electronic drift in the low energy counter, and the data were fitted to the function  $W(\theta) = 1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)$ . The resulting  $A_2$  and  $A_4$  coefficients were then corrected for the geometry of the detectors according to the method of Rose.<sup>2</sup>

For the total "1360-keV" peak the corrected correlation function was found to be

$$W_T(\theta) = 1 + (0.11 \pm 0.03) P_2(\cos\theta) + (0.44 \pm 0.03) P_4(\cos\theta). \quad (1)$$

The linewidth of the coincident peak was then measured and compared to a standard single line. The standard used was the 1409-keV  $\gamma$  transition in  $\text{Sm}^{152}$ . Knowing the energies of the three  $\gamma$  rays of which the "1360-keV" peak is mainly composed, we concluded from the linewidth data that  $20 \pm 10\%$  of the counts in the total peak were due to the 1390-keV transition.

Using this estimate, we divided the peak into three parts, a lower part *A*, a middle part, and an upper part *B*. The correlation functions for the parts *A* and *B* were determined separately, whereas the region in the middle was omitted in the calculation. Part *A* corresponded to the sum of the 1335- and 1345-keV transitions and included about 60% of all counts used for the calculation of  $W_T(\theta)$ . Part *B* consisted only of the upper tail of the composite peak, containing about 6% of the total counts, in order to select the 1390-keV  $\gamma$  ray free of the two other transitions. The resulting correlations, corrected for geometry, were

$$W_A(\theta) = 1 + (0.15 \pm 0.03) P_2(\cos\theta) + (0.52 \pm 0.05) P_4(\cos\theta), \quad (2)$$

<sup>2</sup> M. E. Rose, Phys. Rev. **91**, 610 (1953).

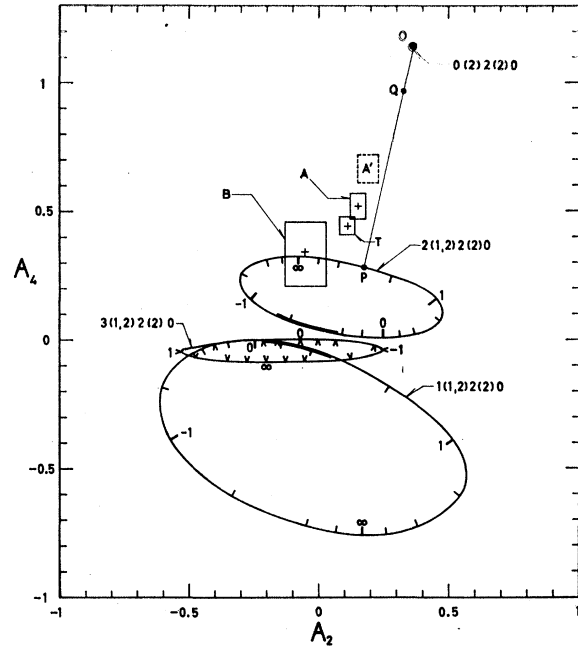


FIG. 2. Parametric plot of  $A_2$  and  $A_4$  for the cascades  $0(2)2(2)0$ ,  $1(1,2)2(2)0$ ,  $2(1,2)2(2)0$ , and  $3(1,2)2(2)0$ . The parameter used is  $\delta$ , the ratio of quadrupole to dipole reduced matrix elements. This type of representation was first suggested by C. F. Coleman [Nuclear Phys. **5**, 495 (1958)], and P. S. Jastram, G. T. Wood, and J. P. Hurley [Bull. Am. Phys. Soc. **3**, 65 (1958)]. The parameter values  $0, \pm 0.1, \pm 0.2, \pm 0.5, \pm 1, \pm 2, \pm 5, \pm 10$ , and  $\infty$  are indicated by marks on the ellipses. The crosses represent the measured  $A_2, A_4$  values and the surrounding rectangles indicate the magnitudes of the experimental uncertainties. The rectangle *T* shows the correlation for the total "1360"-keV peak, and the rectangles *A* and *B* show the correlations for the lower and upper parts of the peak, respectively. Rectangle *A'* represents the possible  $A_2, A_4$  values for the correlation *A* with assumed attenuation coefficients  $G_2 = G_4 = 0.8$ .

$$W_B(\theta) = 1 - (0.05 \pm 0.08) P_2(\cos\theta) + (0.34 \pm 0.13) P_4(\cos\theta). \quad (3)$$

The figures for correlation *A* include a small correction for counts due to the 1390-keV photon.

A graphical representation of the situation is shown in Fig. 2 in which the theoretical correlation coefficients  $A_2$  and  $A_4$  are plotted for various cascades having the spin sequence  $J-2-0$ , where  $J=0, 1, 2, 3$ . According to GNN, these are the only possible spin sequences for the three cascades in question. The experimental limits on  $A_2$  and  $A_4$  are indicated by rectangles which surround the experimental points.

Considering the position of the rectangle *T*, representing the experimental result for the total correlation, it is seen that at least one of the three high-lying states in question must have spin 0. This is consistent with the  $0+$  assignment to the 1440-keV state which was proposed by GNN on the basis of internal conversion data. In the following we shall, therefore, assume the 1345-93 keV cascade to be of the  $0-2-0$  type and fix our attention on the two other cases.

Let us first consider the 1390–93 keV cascade whose properties are determined by the rectangle *B* (Fig. 2). In order to draw detailed conclusions from the position of this rectangle we must know to what extent extranuclear attenuation effects have influenced the correlations. With the source material at our disposal, no experiment seemed feasible which would yield this information directly. In particular, we note that the 214–93 keV 4–2–0 rotational cascade is too weak in the 9.3-min Ta<sup>178</sup> decay to allow a  $\gamma$ – $\gamma$  directional correlation measurement. We can, however, estimate the attenuation by the following argument. Requiring the experimental result for the 1390–93 keV cascade to be consistent with at least one of the theoretical correlation coefficients represented on Fig. 2, we are left only with the possibilities 0 or 2 for the spin of the 1483-keV state. In order to explain the large discrepancy which exists in the spin 0 case between theoretical and experimental correlation coefficients we must assume the attenuation to be extremely large. In terms of attenuation coefficients<sup>3</sup> we find  $G_2 \lesssim 0.1$  and  $G_4 \lesssim 0.3$ , which appears rather improbable in view of the lifetime of the 93-keV state and the chemical environment of the Ta<sup>178</sup> nuclei. A more likely interpretation of the data is that the 1483-keV state has spin 2, in agreement with the tentative 2+ assignment proposed by GNN. With this interpretation it is not necessary to introduce any attenuation to explain the experimental results, and the lower limit which can be put on the attenuation factor  $G_4$  is about 0.65.

Adopting the spin 2 assignment, we see from Fig. 2 that the 1390 keV radiation is mainly quadrupole, the amount of dipole radiation being limited by the inequalities  $\delta \gtrsim 7$  or  $\delta \lesssim -13$ , where  $\delta$  denotes the ratio of quadrupole to dipole reduced matrix elements. This estimate is based on the assumption that  $G_2 = 0$ , but it appears from the diagram that the possible presence of a small attenuation will not affect the result significantly.

We next turn to the 1335–93 keV cascade. The information about the corresponding  $\gamma$ – $\gamma$  angular correlation function must be extracted from the function  $W_A(\theta)$  (the rectangle *A* on Fig. 2) which represents the incoherent sum of the 1335–93 and 1345–93 keV correlations. The analysis is facilitated by the fact that, according to our previous statement, the latter correlation can be represented by the single point *O* in the diagram which corresponds to a 0–2–0 spin sequence. In order to obtain the 1335–93 keV correlation function we must also know the intensity ratio of the 1335- and 1345-keV  $\gamma$  rays; unfortunately, this quantity is not given by experiment. Instead, we base the analysis on the internal conversion data reported by GNN and on their conclusions regarding the spin and parity of the 1430-keV state, which may be summarized as follows. The spin and parity may be either 1+, in which case the 1335-keV photon is an un-

TABLE I. Summary of analysis of 1335–93 keV correlation.

Spin sequence	Assumed multipole mixture <sup>a</sup>	Quadrupole to dipole mixing ratio $\delta$	Relative content of quadrupole radiation
1–2–0	<i>M1</i> + <i>E2</i>	$+0.04 < \delta < 0.3$	$0.2\% \lesssim E2 \lesssim 8\%$
2–2–0	<i>E1</i> + <i>M2</i>	$-0.6 < \delta < -0.2$	$4\% \lesssim M2 \lesssim 27\%$
3–2–0	<i>E1</i> + <i>M2</i>	No acceptable part on the ellipse.	

<sup>a</sup> From GNN.

known *M1*+*E2* mixture, or it may be 2– or 3–, in which cases the decay radiation is an unknown *E1*+*M2* mixture. The data of GNN are also consistent with a 0+ assignment, but this is ruled out by our results, providing the attenuation is small. Thus, according to GNN, the angular correlation of the 1335- and 93-keV photons can be represented by an unspecified point on one of the three ellipses in Fig. 2. Only points on certain parts of the ellipses are, however, compatible with our angular correlation data. These parts may be found by the following geometrical procedure. Let us consider an arbitrary point *P*, say, on the 2–2–0 ellipse. From the mixing ratio  $\delta$  associated with the position of *P*, we calculate the *M2*/*E1* intensity ratio in the 1335-keV radiation, and the corresponding internal conversion coefficient may then be obtained by means of the theoretical *E1* and *M2* internal conversion coefficients. Knowing the intensity of the 1335-keV *K*-conversion line from the measurements of GNN, we can in turn calculate the 1335-keV  $\gamma$ -ray intensity  $I_{\gamma, 1335}$  corresponding to our choice of *P*. The 1345-keV  $\gamma$  ray, on the other hand, is a pure *E2* radiation and, using the theoretical *E2* internal conversion coefficient and the measured *K*-conversion intensity,<sup>1</sup> we can independently establish its  $\gamma$ -ray intensity. We are now in the position to construct the point *Q* in the diagram, which corresponds to the incoherent sum of the two correlations represented by the points *O* and *P*. As is readily seen, *Q* is situated on the line *OP*, dividing it in the ratio  $OQ:PQ = I_{\gamma, 1335}:I_{\gamma, 1345}$ . If *Q* falls inside the rectangle *A*, the particular choice of  $\delta$  for the 2–2–0 spin sequence is compatible with our angular correlation results. If *Q* falls outside of *A*, the corresponding value of  $\delta$  is rejected.

In this analysis we have hitherto assumed the rectangle *A* to represent the true correlation. If some attenuation is present, the true correlation is represented by another rectangle *A'*, displaced correspondingly towards the upper right in the diagram. We have allowed for the possible presence of a reasonably large attenuation by accepting all points *Q* falling inside rectangles *A'*, corresponding to the limits  $0.8 \leq G_2 \leq 1$  and  $0.8 \leq G_4 \leq 1$ . Also, we have taken into account the quoted<sup>1</sup> 20% error on the internal conversion electron intensities, when establishing the  $\gamma$ -ray intensities.

The result of the analysis is summarized in Table I, and the acceptable parts of the ellipses are indicated in heavy print on Fig. 2.

<sup>3</sup> See, e.g., H. Frauenfelder, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 581.

The 3- assignment to the 1430-kev state is ruled out by our results, but an unambiguous choice between the other two possibilities cannot be made on the basis of the present experimental material. It was pointed out by GNN, however, that the low  $\log ft$  value for the beta branch populating this state indicates that most likely the spin and parity is 1+. This assignment is certainly consistent with the angular correlation data and it is interesting to note that in this case the de-excitation to the 2+ rotational state takes place by an almost pure  $M1$  transition. The 2- possibility is also consistent with our data, but it implies the presence of a relatively large amount of  $M2$  radiation in the 1335-kev transition corresponding to a rather strong retardation of the  $E1$  part.

Reviewing the information which has been obtained from the directional correlation data, we observe that these support the more tentative spin assignments which were given by GNN for the 1430- and 1483-kev states. Furthermore, the correlation data have determined the multipolarities of the  $\gamma$  rays which proceed from these states to the 2+ rotational state. This increased amount of information permits us to consider the properties of the 1390-kev transition in more detail than was possible for GNN. We are here dealing with a transition which takes place between two states, both having spin and parity 2+ and, therefore, the transition may proceed by a mixture of  $E0$  and higher order multipole radiations. In this connection it is interesting to note that the transition from the 1277-kev, 2+ level in  $\text{Hf}^{178}$  to the 2+ rotational state according to GNN contains an appreciable  $E0$  admixture.

In order to estimate the amount of  $E0$  radiation in the

1390-kev radiation from the present data we assume that the spin and parity of the 1430-kev state is 1+ and, for simplicity, that the 1335-kev and 1390-kev  $\gamma$  rays are pure  $M1$  and pure  $E2$ , respectively. Using the line-width data, we estimate the intensity of the 1390-kev  $\gamma$  ray to be about 29% of the total intensity of the 1335- and 1345-kev  $\gamma$  rays. The number of monopole  $K$  electrons in the 1390-kev transition divided by the total number of 1390-kev  $K$ -conversion electrons is then given approximately by

$$\frac{I_{e, E0, 1390}}{I_{e, E0+E2, 1390}} = 1 - 0.29 \left( \frac{I_{e, 1335}}{\beta_{1, 1335}} + \frac{I_{e, 1345}}{\alpha_{2, 1345}} \right) \frac{\alpha_{2, 1390}}{I_{e, 1390}} \approx 0.7. \quad (4)$$

Here,  $\alpha_2$  and  $\beta_1$  are theoretical  $K$ -conversion coefficients and  $I_{e, 1335}$ ,  $I_{e, 1345}$ , and  $I_{e, 1390}$  denote measured  $K$  electron intensities taken from the work of GNN.

Due to the large uncertainty involved in the analysis of the broad "1360-kev" peak, the above calculation represents only a rough estimation, but it clearly shows that a considerable amount of the 1390-kev  $K$  electrons is due to electric monopole transitions. Thus, it seems that at least two of the 2+ to 2+ transitions in  $\text{Hf}^{178}$  have  $E0$  components.

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