Angular Correlations of Gamma Rays in Ta^{181}

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The directional angular correlation of three different gamma-ray cascades in Ta^{181} have been measured with a fast delayed-coincidence scintillation spectrometer employing NaI phosphors as detectors. An interpretation of the results, in combination with measured K-shell internal-conversion coefficients, indicates that an unambiguous assignment of the angular momenta of three excited states in Ta^{181} and a classification of the gamma rays are possible. The spin sequences are as follows: for the 132-480 kev gamma-ray cascade $5/2(E2)9/2(E2+M1)7/2$ with δ_{480}, the square root of the intensity ratio of electric quadrupole to magnetic dipole radiation, equal to -1.25; for the 132–345 kev gamma-ray cascade $5/2(E2)9/2(E2+M1)9/2$ with $\delta_{345}=1.0\pm0.2$; and for the 345–135 kev gamma-ray cascade $9/2(E2+M1)9/2(E2+M1)7/2$ with $\delta_{345}=0.95\pm0.10$ and $\delta_{135}=0.5\pm0.05$. The observed angular correlation functions for the 132–480 kev and 132–345 kev cascades indicate an appreciable attenuation by perturbing interactions in the intermediate state of the nucleus by its surroundings, even in the liquid state. For polycrystalline hafnium metal and hafnium compounds, the attenuation coefficients are smaller than the "hard core" values for a static electric quadrupole interaction.

I. INTRODUCTION

THE radiations from the decay of Hf^{181} have been studied by many investigators.[1] In spite of the numerous activities produced by neutron-irradiated Hf, the general features of the decay scheme of Hf^{181} given originally by Chu and Wiedenbeck[2] are probably correct.[3] Several different sets of spins have been assigned the excited states in Ta^{181} on the basis of relative gamma-ray intensities, lifetimes, K/L ratios, and internal-conversion electron data.[4–6] It is apparent that the data and assignments are not all consistent. In the level assignments by Goldhaber and Sunyar[5] and Fan,[6] nearly all the spins ($d_{5/2}$, $d_{3/2}$, and $s_{1/2}$) predicted by the nuclear shell model were included. As we shall see from the interpretation of the results in this paper, the primitive version of the nuclear-shell model has practically no value for predicting level assignments in Ta^{181}. Fan concluded that all of the gamma-ray transitions were pure multipoles on the basis of internal conversion coefficients obtained from electron intensity measurements. These conclusions are not in accord with some recent measurements of the K-shell internal-conversion coefficients of the gamma rays in Ta^{181}.[7]

In many nuclear gamma transitions, a measurement of K-shell internal-conversion coefficients will permit an unambiguous assignment of multipole order and character of the transition. However, in some cases of a mixed multipole transition, a measured conversion coefficient will not distinguish between a parity-favored or a parity-unfavored transition.[7] If these measurements are supplemented by directional angular correlation measurements, it should be possible to infer, in addition to the spins of the states, both the character of the gamma rays and relative parity of the states. In this paper, the application of a fast delayed coincidence scintillation spectrometer to the measurement of the directional angular correlation of three different gamma-ray cascades in Ta^{181} is described. The experimental measurements and results are presented. Finally, an interpretation of the results, in combination with the K-shell internal-conversion coefficients,[7] is discussed.

II. APPARATUS

For the detection of the radiations, scintillation detectors consisting of thallium-activated sodium iodide crystals mounted on RCA type 5819 photomultiplier tubes were used. The lateral sides and front face of the crystals were covered with 30 mg/cm^2 of MgO and 0.010 in. of Al.[8] Fluorothene source holders with 0.025-in. wall thickness were located at the intersection of the axes of the cylindrical crystals at a distance of 7 cm from the front face of each crystal. The delayed coincidence scintillation spectrometer which uses the fast-slow coincidence method has been described previously.[7] The windows of the differential pulse-height analyzers of the coincidence spectrometer in the angular correlation apparatus are always set to include only the full-energy pulse spectrum peak of the gamma ray. Most of the data in these experiments were recorded automatically at 90°, 135°, 180°, 225°, and 270°. The time for the collection of a fixed number of coincidence counts was printed on a paper tape by a printing timer, but the angular positions were changed manually.

III. EXPERIMENTAL MEASUREMENTS AND RESULTS

Sources of Hf^{181} were prepared from samples of Hf metal and HfO_2 irradiated with pile neutrons for four

[1] *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950) and Supplements 1, 2, and 3. "New Nuclear Data 1952 Cumulation," Nuclear Science Abstracts 6, No. 24B (1952).
[2] K. Chu and M. Wiedenbeck, Phys. Rev. 75, 226 (1949).
[3] A. Hedgran and S. Thulin, Phys. Rev. 81, 1072 (1951).
[4] W. C. Barber, Phys. Rev. 80, 332 (1950).
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[8] C. J. Borkowski, Rev. Sci. Instr. 24, 1046 (1953).

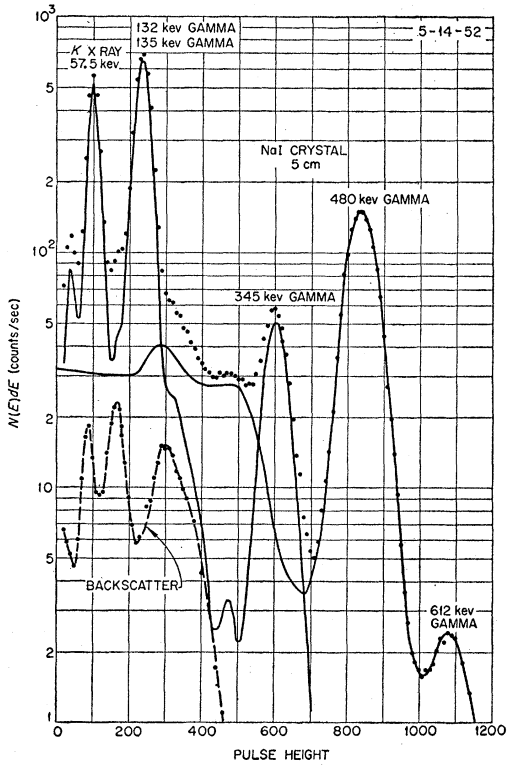

FIG. 1. A differential pulse height spectrum of the gamma radiation from $Hf^{181} \rightarrow Ta^{181}$.

weeks. The sample of HfO_2 was enriched in Hf^{180} (93.96 percent).[9] With this enrichment, the intensity of the 342-kev gamma ray of Hf^{175} in the source relative to the 345-kev gamma ray of Hf^{181} is about 4 percent. With sources of normal Hf, the intensity of the 342-kev gamma ray is equal to the 345-kev gamma-ray intensity three weeks after the end of the irradiation. A differential pulse-height spectrum of the gamma radiation from $Hf^{181} \rightarrow Ta^{181}$ is shown in Fig. 1.

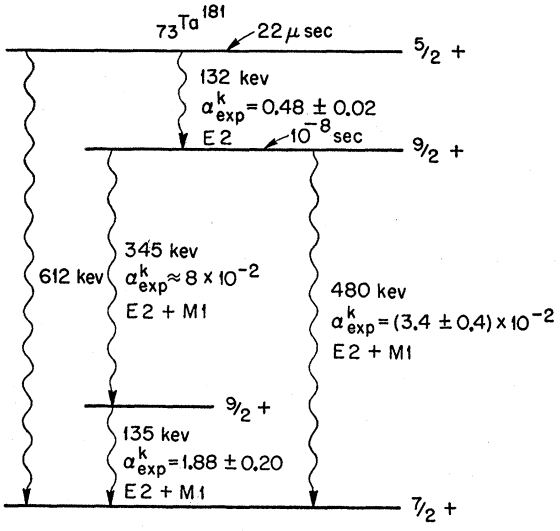

FIG. 2. Decay scheme of Ta^{181} with the spin assignments of the excited states and the character of the gamma rays.

[9] Obtained from the Isotopes Division of the Oak Ridge National Laboratory.

A. 132–480 kev Cascade

Since the intermediate state of this cascade (see Fig. 2) is metastable with a half-life of 10^{-8} sec, the initial experiments were devoted to measurements of the anisotropy, $A' = [C(\pi)/C(\pi/2)] - 1$, as a function of the chemical and physical state of the source, where $C(\pi)$ and $C(\pi/2)$ are the coincidence counting rates at $\theta = \pi$, and $\pi/2$, respectively. For this cascade, sources prepared from normal Hf may be used because coincidences from the Hf^{175} gamma-ray cascades are not detected under the conditions for which the data were collected. Since a resolving time, $2\tau_0$, of 0.18 μsec was used, the measurements represent the anisotropy of the integral angular correlation, i.e., all of the 480-kev gamma rays are detected with equal probability with regard to the time of emission of the gamma ray. In Table I, the anisotropy, A', of the 132–480 kev gamma cascade in Ta^{181} with different sources of Hf^{181} is summarized. Since Hf is a rather difficult element to work with from the standpoint of chemistry, only a few compounds were studied. The largest anisotropy found is exhibited by solution sources of Hf metal and HfO_2 taken up in concentrated HF as a solvent. Unless otherwise specified, this type of source was used in all experiments to be discussed in this paper.

A series of experiments was performed to measure the coefficients A_2 and A_4 of the terms in the expansion of the correlation function in Legendre polynomials for the 132–480 kev gamma-ray cascade. These measurements included both the integral and differential directional angular correlation, i.e., directional angular correlation as a function of the time of emission of the 480-kev transition. The operating conditions and the angular correlation coefficients, corrected for finite angular resolution,[10] are tabulated in Table II. The true coincidence counting rate for this cascade was of the order of 3 counts per sec while the random rate is about 15 percent and 1 percent of this for $2\tau_0 = 0.18$ μsec and 1.32×10^{-8} sec, respectively. From the results in Table II the anisotropy in the angular correlation of the 132–340 kev gamma-ray cascade is independent of the emission time of the 480-kev transition to within ± 3.2 percent.

B. 132–345 kev Cascade

To obtain the experimental angular correlation function for the 132–345 kev gamma-ray cascade, the coincidences were collected at a time delay of 10^{-8} sec, which is sufficient to resolve all prompt coincidences due to the 345–135 kev gamma-ray cascade. Only sources of Hf^{181} prepared from irradiated samples of HfO_2

[10] M. E. Rose, Phys. Rev. **91**, 610 (1953).

enriched in Hf^{180} were used in these measurements. The window of one differential pulse height analyzer, which accepted only the full-energy pulse-spectrum peak of the 345-kev gamma ray, also accepted a small portion of the Compton recoil electron pulse distribution from the 480-kev gamma ray. Thus, the data collected represented a composite correlation function of the 132–480 kev and 132–345 kev gamma-ray cascades. The contribution from the 132–480 kev gamma-ray cascade was removed from the composite data by a direct measurement of the 132–480 kev gamma-gamma correlation at the same time delay and by a determination of the number of counts in the window of the

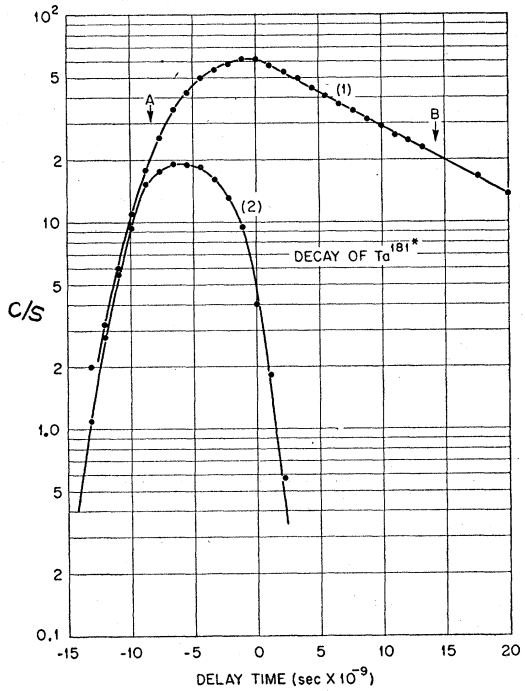

FIG. 3. Curve (1) is the number of coincidences as a function of the time delay obtained between the 132-kev and 480-kev gamma rays of Ta^{181} which represents the decay of Ta^{181*} (10^{-8} sec). Curve (2) is a time-resolution curve for prompt events obtained between annihilation gamma rays from Na^{22}.

analyzer from the 480-kev transition. A procedure for constructing the shape and intensity of the Compton recoil electron pulse distribution of the 480-kev gamma radiation from a differential pulse-height spectrum of the 512-kev gamma radiation from Sr^{85} has been described previously.[7] In this way and with a knowledge of the window width, the number of counts in the window from the 480-kev transition was obtained by (a) an integration of the differential pulse-height spectrum over the width of the window used in the angular correlation measurements and (b) a direct measurement of the counts in window with a source of Sr^{85} corrected for the difference in the intensities of the two Compton distributions.[7] Good agreement was

TABLE I. Effect of chemical and physical state of Hf sources on the angular correlation of the 132-480 kev gamma-ray cascade.

Sample	Solvent	Form of the source	Anisotropy $A'=\frac{C(\pi)}{C(\pi/2)}-1$
Hf metal		Polycrystalline Hf	$-(0.06\pm0.01)$
Hf metal	27*N* HF	0.10 ml of solution	$-(0.38\pm0.01)$
Hf metal	27*N* HF	0.10 ml of solution plus 0.10 ml $HClO_4\cdot 2H_2O$	$-(0.38\pm0.02)$
Hf metal	27*N* HF	Above source 24 hr after preparation	$-(0.31\pm0.02)$
Hf metal	27*N* HF	Above source 48 hr after preparation	$-(0.26\pm0.02)$
Hf metal	27*N* HF	Above source 21 days after preparation	$-(0.22\pm0.02)$
Hf metal	0.27*N* HF	0.10 ml of solution	$-(0.32\pm0.02)$
HfF_4	12*N* HCl	0.10 ml solution of $HfOCl_2$	$-(0.28\pm0.01)$
Hf metal		Hf extracted into a benzene layer containing TTA	$-(0.12\pm0.01)$
$Hf(OH)_4$	8*N* HNO_3	Gelatinous	$-(0.10\pm0.01)$
$Hf(OH)_4$		Dry	$-(0.09\pm0.01)$
$Hf(NO_3)_4$	8*N* HNO_3	0.02 ml of solution	$-(0.19\pm0.02)$
HfO_2	27*N* HF	0.02 ml of solution	$-(0.38\pm0.01)$
HfO_2		Polycrystalline HfF_4 and $HfOF_2$	$-(0.08\pm0.01)$

obtained between the two measurements. The composite coincidence rate was of the order of 1 count per sec while the random rate was about 3 percent of this. The contribution from the 132–480 kev cascade to the composite coincidence rate was between 20 and 30 percent of this. The results from the measurements are given in Table II.

C. 345–135 kev Cascade

In order to get a measurement of the angular correlation function of the 345–135 kev gamma-ray cascade, the following set of experiments was performed. At a time delay A of Fig. 3, coincidences were collected resulting from the 132–480 kev, 132–345 kev, and

TABLE II. Directional angular correlation coefficients for the 132–480 kev and 132-345 kev gamma cascades in Ta^{181}.

Cascade	Operating conditions	A_2	A_4
132–480 kev	$2\tau_0=0.18$ μsec	−0.2789	−0.0565
132–480 kev	$2\tau_0=0.18$ μsec	−0.2747	−0.0554
132–480 kev	$2\tau_0=0.18$ μsec	−0.2820	−0.0587
132–480 kev	$2\tau_0=0.18$ μsec	−0.2747	−0.0554
132–480 kev	$2\tau_0=1.32\times10^{-8}$ sec; time delay $=6\times10^{-9}$ sec	−0.2765	−0.0700
132–480 kev	$2\tau_0=1.32\times10^{-8}$ sec; time delay $=6\times10^{-9}$ sec	−0.2622	−0.0524
132–480 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	−0.2866	−0.0487
132–480 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	−0.2787	−0.0664
132–480 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	−0.3061	−0.0623
132–345 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	0.1306	0.0045
132–345 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	0.1081	0.0529
132–345 kev	$2\tau_0=1.40\times10^{-8}$ sec; time delay $=10^{-8}$ sec	0.0909	0.0303

345–135 kev gamma-ray cascades. Since the observed angular correlation functions for the 132–480 kev and 132–345 kev gamma-ray cascades are independent of the time of emission of the 480- and 345-kev transitions, their effect in the composite correlation function was was removed by a direct measurement. That is, the coincidence rates for the 132–480 kev and 132–345 kev gamma-ray cascades were measured at a time delay B. At this time delay, these coincidence rates are equal to the contributions of the 132–480 kev and 132–345 kev cascades to the coincidence rates at time delay A. A determination of the time delays A and B used in these measurements was obtained from a measurement of time resolution curves for both prompt and delayed events. For instance, these latter experiments involved a measurement of a shift of 6×10^{-10} sec of the centroids of two time resolution curves to within $\pm5\times10^{-11}$ sec. In Fig. 3, curve (1), the number of coincidences are plotted as a function of the delay time obtained with a source of Hf^{181}. This delayed-coincidence resolution curve was recorded with the window of one analyzer accepting only the full-energy pulse spectrum peak of the 132-kev gamma ray and the window of the other analyzer accepting only the full-energy pulse spectrum peak of the 480-kev gamma ray. Without a change in the apparatus, a resolution curve for prompt events (curve 2) was obtained between annihilation gamma rays from a source of Na^{22}. However, in the angular correlation measurements the window of one analyzer included only the full-energy pulse spectrum peak of the 345-kev gamma ray. A second time resolution curve for prompt events with the window set for pulses corresponding to 345 kev was taken (not shown in Fig. 3). This curve was shifted 6×10^{-10} sec towards positive time delays with respect to curve (2) of Fig. 3. Thus, a delayed coincidence resolution curve of 132–480 kev and 132–345 kev coincidences with the window of the analyzer on the full-energy pulse spectrum peak of the 345-kev gamma ray would be shifted a like amount. In the angular correlation measurements the time delays A and B corresponded to -7.7 and 15.4×10^{-9} sec, respectively.

In addition to the contribution from the 132–480 kev and 132–345 kev gamma-ray cascades, there was another source of coincidences at a time delay of -7.7×10^{-9} sec which was not present at a time delay of 15.4×10^{-9} sec arising from the Compton scattering of the 480-kev gamma ray between the detectors. This effect was small because the position and width of the windows of the analyzers discriminated almost completely against such a source of coincidences. However, this source of coincidences was measured directly using the 512-kev gamma ray from Sr^{85}. The coincidence rate was an isotropic 10 percent effect and was removed from the composite data.

To check the influence of perturbing interactions in the intermediate state by extranuclear fields on the angular correlation of the 345–135 kev gamma-ray cascade, the measurements were taken with a solution source of HfO_2 in hydrofluoric acid and with a polycrystalline source having a probable composition of HfF_4 and $HfOF_2$. This polycrystalline source was prepared from the same solution as was used in the liquid source by evaporating the solution to dryness in a fluorothene source holder under a heat lamp. Identical angular correlation functions were obtained, indicating that the intermediate state is prompt or that the perturbing interactions are small compared to those observed in the 132–480 kev gamma-gamma angular correlation. This result also assures one that the contributions from the 132–480 kev and 132–345 kev gamma-ray cascades have been removed from the composite data properly because the contributions from these two cascades are quite different for liquid and polycrystalline sources (see Table I). The angular correlation coefficients, corrected for finite angular resolution,[10] for the 345–135 kev gamma-ray cascade are tabulated in Table III. The contribution of the 132–480 kev and 132–345 kev coincidences to the composite coincidence rates was about 20 percent, where the composite rates were of the order of 1.5 counts per sec.

TABLE III. Directional angular correlation coefficients for the 345–135 kev gamma-ray cascade in Ta^{181}.

Form of the source	Experimental Coefficients A_2	A_4
$Hf^{181}O_2$ in 27N HF as a solvent	0.2003	−0.0343
$Hf^{181}O_2$ in 27N HF as a solvent	0.2292	−0.0541
Polycrystalline HfF_4 and $HfOF_2$	0.2010	−0.0962
Polycrystalline HfF_4 and $HfOF_2$	0.2196	−0.0712
$Hf^{181}O_2$ in 27N HF as a solvent	0.1908	−0.0547
Polycrystalline HfF_4 and $HfOF_2$	0.1710	−0.0086

TABLE IV. Mean values of the angular correlation coefficients for the gamma-ray cascades in Ta^{181}.

Cascade	Number of coincidences collected	Experimental coefficients A_2	A_4	Anisotropy $A=\frac{W(\pi)}{W(\pi/2)}-1$
132–480 kev	4.3×10^6	$-(0.280\pm0.004)$	$-(0.058\pm0.003)$	−0.408
132–345 kev	6.8×10^5	0.110 ± 0.012	0.029 ± 0.014	0.192
345–135 kev	1.6×10^6	0.202 ± 0.009	$-(0.053\pm0.014)$	0.295

IV. INTERPRETATION OF RESULTS

In Table IV the directional angular correlation coefficients for the gamma-ray cascades in Ta^{181} are summarized. An analysis of these results, in combination with the measured K-shell internal conversion coefficients[7] shown in Fig. 2, indicates that an unambiguous assignment of the angular momenta of these excited states in Ta^{181} and a classification of these

gamma rays are possible. Many spin sequences must be considered in the analysis and for completeness those that are excluded by the experimental results are enumerated. In the case of the 132–480 kev gamma-ray cascade, none of the following spin sequences fit experiment: $j\pm2(E2+M3)j=7/2$, $7/2\pm1(Q+D)\times7/2$; j or $j\pm1(Q+D)j=7/2$, $7/2\pm1(Q+D)7/2$; j, $j\pm1$, or $j\pm2(E2)j=7/2$, $7/2\pm1(Q+D)7/2$, except the decay sequence $5/2(E2)9/2(E2+M1)7/2$. In this notation $j_1=j$, $j\pm1$, or $j\pm2$ and all combinations are considered. The unperturbed angular correlation coefficients,[11] A_2 and A_4, are shown in Fig. 4 as a function of $|\delta_2|$, where δ_2[2] is the ratio of squares of the reduced matrix elements and is defined as the intensity ratio (in this case) of quadrupole to dipole radiation in the 480-kev transition. The experimental values of A_2 and A_4 and the value of δ_{480} obtained from the K-shell internal-conversion coefficient on the assumption that the transition is $E2+M1$ radiation are plotted in Fig. 4. A mixture of $E1+M2$ radiation is excluded by the fact that $(A_4)_{\text{exp}}$ is larger than the unperturbed A_4. These results indicate an appreciable attenuation in the directional angular correlation coefficients. The perturbed angular correlation function can be written as

$$W(\theta)=1+\sum_\nu G_{\nu j}A_\nu P_\nu(\cos\theta),$$

where $G_{\nu j}$ are the attenuation coefficients which represent the effect of perturbing interactions in the intermediate state of the nucleus. These attenuation coefficients are independent of the nature of the transitions to and from the intermediate state.[12] For $\delta_{480}=-1.25$, the experimental attenuation coefficients are $G_{2,9/2}=0.63$ and $G_{4,9/2}=0.60$. If these attenuation coefficients are attributed entirely to the time-dependent interaction[13] of the nuclear electric quadrupole moment of the 480-kev state with the gradient of the electric field from its surroundings (liquid state), these values are not too consistent. However, for $\delta_{480}=-1.7$, a consistent set of attenuation coefficients, $G_{2,9/2}=0.735$ and $G_{4,9/2}=0.497$, is obtained. This leads to $\alpha^K(480\text{ kev})=2.8\times10^{-2}$, which is still in fair agreement with the measured value.

From this consistent set of attenuation coefficients for $\delta_{480}=-1.7$, one may make an estimate of the electric quadrupole interaction energy.[13] For $\tau_N=1.5\times10^{-8}$ sec, and if the correlation time τ_c is taken to be 10^{-11} sec, then $eQ(\partial^2V/\partial z'^2)/h$ is 1949 Mc/sec. If the electric quadrupole moment of the 480-kev state in Ta^{181} is comparable to the ground-state quadrupole moment of 6×10^{-24} cm^2, which is one of the largest quadrupole moments known, than this quadrupole interaction is probably by no means unreasonably large.

Since many spin sequences are already excluded by the 132–480 kev angular correlation function, only two

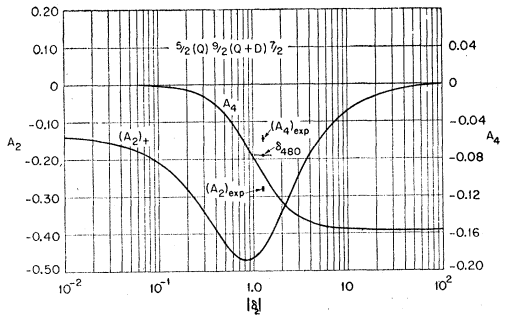

FIG. 4. Unperturbed angular correlation coefficients as a function of $|\delta_2|$ for the decay sequence $5/2(Q)9/2(Q+D)7/2$. The curve labelled $(A_2)_+$ corresponds to taking δ_2 positive. *Note added in proof.*—The curve labelled $(A_2)_+$ should read $(A_2)_-$ and corresponds to taking δ_2 negative. The change of sign is due to a misprint in Eq. (70c) of a prepublication copy of the paper referred to in reference 11.

spin sequences need be considered in the analysis of the 132–345 kev angular correlation data. These are $5/2(E2)9/2(Q+D)9/2$ and $5/2(E2)9/2(Q+D)\times7/2$. The sign of the coefficient $(A_4)_{\text{exp}}$ immediately excludes the latter spin sequence. The unperturbed angular correlation coefficients, A_2 and A_4, as a function of $|\delta_2|$ are shown in Fig. 5. The attenuation coefficients determined from the 132–480 kev angular correlation data should, of course, be the same in the 132–345 kev angular correlation and are used in the analysis to determine δ_{345}. Unperturbed experimental coefficients, $(A_\nu)_{\text{exp}}/G_{\nu,9/2}$, for $\delta_{480}=-1.25$ are plotted in Fig. 5. From the value of $(A_2)_{\text{exp}}/G_{2,9/2}$, one finds $\delta_{345}=0.93$. If the consistent set of attenuation coefficients corresponding to $\delta_{480}=-1.7$ is used, δ_{345} is equal to 1.08. A final value for δ_{345} determined from the 132–345 kev angular correlation data is 1.0 ± 0.2. This value is in good agreement with the absolute value of δ_{345} obtained from the K-shell internal-conversion coefficient on the assumption that the transition is $E2+M1$ radiation.

Although $\alpha^K{}_{\text{exp}}(135\text{ kev})$ indicates that the transition is predominantly $M1$, the uncertainty in the measurement does not exclude a small admixture of $E2$ radiation. However, the presence of a $P_4(\cos\theta)$ term in the

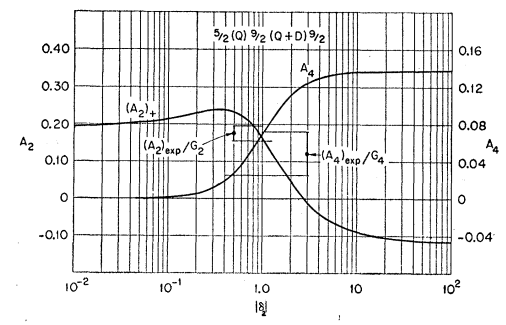

FIG. 5. Unperturbed angular correlation coefficients as a function of $|\delta_2|$ for the decay sequence $5/2(Q)9/2(Q+D)9/2$. The curve labelled $(A_2)_+$ corresponds to taking δ_2 positive.

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[12] Kurt Alder, Helv. Phys. Acta **25**, 235 (1952).
[13] A. Abragam and R. V. Pound, Phys. Rev. **92**, 936 (1953).

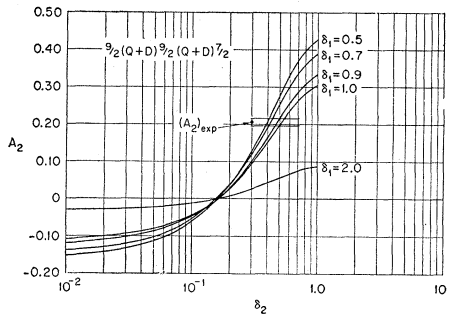

FIG. 6. Unperturbed angular correlation coefficient A_2 as a function of δ_2 for various values of δ_1 for the decay sequence $9/2(Q+D)9/2(Q+D)7/2$.

observed angular correlation function of the 345–135 kev gamma-ray cascade requires an admixture of $E2$ radiation. This is the first correlation which has been observed in which each transition is a mixture of electric and magnetic multipole radiation. An explicit form of the directional angular correlation function for a mixed-mixed gamma-ray cascade has been derived by Rose.[14] A computation of the angular correlation coefficients for a mixed-mixed gamma-ray cascade for all possible values of δ_1 and δ_2 and many possible spin sequences is quite tedious. However, in this case of the 345–135 kev gamma-ray cascade the coefficient A_4 is independent of the sign of δ_1 and δ_2 and $\alpha^K{}_{\text{exp}}$(135 kev) restricts $|\delta_2|\leq 0.5$. As a result many of the possible spin sequences, j, $j\pm 1(Q+D)j=7/2$, $7/2\pm 1(Q+D)\times 7/2$, that need be considered are excluded by examining the sign of the coefficient A_4. Only the spin sequence $9/2(E2+M1)9/2(E2+M1)7/2$ fits experiment. The angular correlation coefficients, A_2 and A_4, are shown as a function of δ_1 and δ_2 in Figs. 6 and 7, respectively. From the 345–135 kev angular correlation another independent determination of δ_{345} is obtained which is in good agreement with the value from the analysis of the 132–345 kev angular correlation data.

A summary of the interpretations obtained from the analysis of the angular correlation results, in combination with the measured K-shell internal-conversion coefficients, is given in Table V.

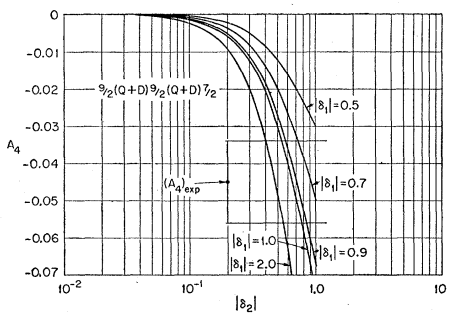

FIG. 7. Unperturbed angular correlation coefficient A_4 as a function of $|\delta_2|$ for various values of $|\delta_1|$ for the decay sequence $9/2(Q+D1)9/2(Q+D)7/2$.

[14] M. E. Rose, following paper [Phys. Rev. **93**, 477 (1954)].

Since we know within limits the unperturbed angular correlation function for the 132–480 kev gamma-ray cascade, it is of interest to examine the magnitude of the anisotropies quoted in Table I for polycrystalline Hf metal and polycrystalline HfF_4 and $HfOF_2$. The unperturbed an isotropy for $\delta_{480}=-1.25$ and -1.70 is -0.6092 and -0.5622, respectively. The "hard core" limits[13] of the attenuation coefficients for a static quadrupole interaction with an electric field having axial symmetry and with no degeneracies would lead to predicted anisotropies of -0.1335 and -0.1185 for $\delta_{480}=-1.25$ and -1.70, respectively. Taking into account the finite angular resolution of the detectors, these values become -0.1212 and -0.1069 which may be compared directly with anisotropies A' (not corrected for finite angular resolution) in Table I. The experimental values are definitely smaller than the "hard core" limit, which is a lower limit because the G_ν(lim) can be larger than $1/(2\nu+1)$ when there are degeneracies or when there are fields of lower than axial symmetry.[13]

TABLE V. Spin assignments of the excited states and character of the gamma-rays in Ta^{181}.

Cascade	Spin sequence	δ_γ
132–480 kev	$5/2(E2)9/2(E2+M1)7/2$	$\delta_{480}=-1.25^{+0.29}_{-0.22}$
132–345 kev	$5/2(E2)9/2(E2+M1)9/2$	$\delta_{345}=1.0\pm 0.2$
345–135 kev	$9/2(E2+M1)9/2(E2+M1)7/2$	$\delta_{345}=0.95\pm 0.1$ $\delta_{135}=0.50\pm 0.05$

V. CONCLUSIONS

The ambiguity[7] that existed in the classification of the 345-kev and 480-kev gamma rays of Ta^{181} from the measured K-shell internal-conversion coefficients has been removed by the directional angular correlation measurements. Although several of the gamma rays are mixed multipoles and only perturbed angular correlation functions were observed for cascades via the 10^{-8} sec intermediate state, it appears that there exists an unambiguous assignment of the spins and relative parties of the excited states in Ta^{181}. This spin sequence is certainly different from those suggested by Goldhaber and Sunyar,[5] Goldhaber and Hill,[15] and Fan[6] in terms of the nuclear shell model. At least for Ta^{181}, the shell model could be very misleading if one considered only spin sequences covered by the shell model in the interpretation of the results presented above.

The $\log_{10}$ (ft) value of the beta transition from $_{72}Hf^{181}$ to the 22-μsec state in Ta^{181} is 7.2.[16] An assignment of $p_{3/2}$ for the ground state of $_{72}Hf^{181}$ is inferred by the nuclear-shell model which has met with considerable

[15] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

[16] A. M. Feingold, Revs. Modern Phys. **23**, 10 (1951).

success for ground state assignments. This assignment is consistent with a first forbidden transition as suggested by the $\log_{10}(ft)$ value.

The observed angular correlation functions for the cascades via the 10^{-8} sec intermediate state indicate an appreciable attenuation by perturbing interactions in the intermediate state of the nucleus by its surroundings even in the liquid state. Also, the measured anisotropy of the 132–480 kev angular correlation for polycrystalline Hf metal and Hf compounds indicates that the attenuation coefficients are smaller than the "hard core" values for a static electric quadrupole interaction.

Since the expected unperturbed angular correlation functions are known with fair certainty, the cascades 132–480 kev and 132–345 kev provide a good case in which to study the influence of electric and magnetic fields on angular correlations. The coefficients of $P_2(\cos\theta)$ and $P_4(\cos\theta)$ for both cascades are reasonably large so that all attenuation coefficients can be measured with reasonable precision. For the cascade in Cd^{111}, which follows the decay of In^{111}, the unperturbed coefficient of $P_4(\cos\theta)$ is rather small ($A_4=-0.001$). However, Hf^{181} possesses the disadvantage that Hf compounds are difficult to prepare.

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Mixed Gamma-Mixed Gamma Angular Correlation

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The γ-γ correlation in which both radiations are mixed, with arbitrary admixture ratios, is discussed.

RECENT experimental evidence indicates that the occurrence of mixed γ radiation (E_L+M_{L+1} or M_L+E_{L+1}) is not uncommon.[1] There are also several cases in which the intensity ratio δ^2 of 2^{L+1} pole to 2^L pole is of order unity (δ^2 neither $\ll 1$ nor $\gg 1$). Therefore it should not be surprising if a γ-γ cascade occurs in which both radiations are mixed. Indeed, the 345–135 kev cascade in Ta^{181} appears to be such a case[2] (both radiations $E2+M1$) and for both $\delta^2\sim 1$. For this reason it seems worth while to give the correlation function applicable in such cases.

The general theory of angular correlation covers the case of an arbitrary γ-γ cascade, and the required correlation function can be obtained from the general formula given by several authors.[3] For the cascade $j_1(L_1,L_1')j(L_2,L_2')j_2$ with δ_i^2 equal to the intensity ratio of $2^{L_i'}$ to 2^{L_i} poles, the correlation function is

$$W(\vartheta)=\sum_\nu P_\nu(\cos\vartheta)\{F_\nu(L_1j_1j)+\delta_1^2F_\nu(L_1'j_1j)$$
$$+2\delta_1(-)^{j_1-j-1}[(2j+1)(2L_1+1)(2L_1'+1)]^{\frac{1}{2}}G_\nu(L_1L_1'j_1j)\}$$
$$\times\{F_\nu(L_2j_2j)+\delta_2^2F_\nu(L_2'j_2j)+2\delta_2(-)^{j_2-j-1}$$
$$\times[(2j+1)(2L_2+1)(2L_2'+1)]^{\frac{1}{2}}G_\nu(L_2L_2'j_2j)\}. \quad (1)$$

The normalization is to $\langle W(\vartheta)\rangle_{Av}=(1+\delta_1^2)(1+\delta_2^2)$. In practice $L_1'=L_1+1$ and $L_2'=L_2+1$. The coefficients F_ν and G_ν are defined in BR [Eqs. (69b) and (70d)] and are tabulated in Tables I and II of that reference. Of course, when spin coupling is present the correlation function is modified in the usual way by inserting attenuation factors Q_ν which are independent of the properties of the emitted radiations.[4]

The presence of two adjustable intensity ratios (the signs of δ_1 and δ_2 may be $\pm$) makes the case under consideration somewhat overly flexible. However, it is quite likely that the study of the mixed-mixed correlation will be useful in conjunction with other experimental data which serve to fix one of the δ's (or δ^2's). For example, there may be internal conversion data for one of the transitions or, as in the case of Ta^{181}, one of the radiations participates in another cascade the correlation for which can be measured separately.[2]

[1] The evidence has been summarized by R. M. Steffen, Indiana Conference on Nuclear Spectroscopy and the Shell Model (unpublished).

[2] F. K. McGowan, preceding paper [Phys. Rev. **93**, 471 (1954)].

[3] For small admixture ratios the explicit correlation function was given by S. P. Lloyd, Phys. Rev. **83**, 716 (1951). For the more general case one obtains the desired result by using, for example, Eq. (64) in L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953), referred to as BR. We use the notation of this reference. Further references to the general formulation are to be found in that paper.

[4] K. Alder, Helv. Phys. Acta **25**, 235 (1952); A. Abragam and R. V. Pound, Phys. Rev. **92**, 936 (1953). These authors denote the attenuation coefficients by G_ν.