Multilevel Analysis of the Total Neutron Cross Section of Pu^{241} below 12 ev*

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Measurements of the total cross section of Pu^{241} have been analyzed with the use of a multilevel formula, under the assumption that the observed resonance asymmetries are due to interference in a small number of fission channels. As was the case for U^{233}, the analysis is interpreted as evidence that there are differences in the average fission widths of resonances belonging to the two possible spin states of the compound nucleus.

I. INTRODUCTION

THE total neutron cross section of Pu^{241} has been measured[1] from 0.02 ev to 2 kev with the Materials Testing Reactor (MTR) fast chopper.[2] In the energy region from 0.02 to 12 ev, the resonance structure is sufficiently well resolved to permit shape analysis of the data. The cross section of Pu^{241} shows a strong fission component,[3] and is thus expected to exhibit much the same resonance characteristics as the other fissionable nuclei, U^{233}, U^{235}, and Pu^{239}. For these nuclei, strong asymmetries have been observed in the resonance shapes, which are attributed to interference in fission.[4–6] Pronounced asymmetries are observed in several of the resonances of Pu^{241} which cannot be described by the single level Breit-Wigner formula. It is reasonable to assume that these asymmetries are evidence for interference in fission. A multilevel formula which adequately describes such interference has been developed by Reich and Moore,[7] and has been used in the analysis[4] of the slow neutron cross sections of U^{233}. In the present analysis, the same assumption was made which had been found to obtain for U^{233}: that fission proceeds through either one or two channels. This assumption is supported by the observed large fluctuation in the sizes of the fission widths, which according to Porter and Thomas,[8] implies that the number of channels available to the process is small.

II. EXPERIMENTAL DATA

The total neutron cross section data shown in Fig. 1 were calculated from transmission measurements on enriched PuO_2 samples.[9] Instrumental resolutions of 0.05 to 2.4 μsec/meter were used. The experimental arrangement has been discussed in detail.[2,10] Because of the wide variation in cross section as a function of neutron energy, two different nominal sample thicknesses were used, 527 barns/atom and 1042 barns/atom. The thinner sample was used particularly over the 0.264 ev resonance where the transmission was too close to zero for the thick sample. Since Pu^{241} beta decays to Am^{241} with a half-life of 13 years, the buildup of Am^{241} into the sample had to be taken into consideration. The effects of the Pu^{239}, Pu^{240}, Pu^{242}, Am^{241} and oxygen contaminants were removed from the raw data in order to obtain the Pu^{241} total cross section. The data were corrected for the contaminant cross sections except in the regions where resonances appeared in the contaminant data. Above 1 ev, resonance structure due to contaminants was not removed because of differences in the instrumental resolution and sample thickness between these Pu^{241} measurements and the measurements reported for the contaminants.[11] All cross section effects due to the contaminant resonances have been removed below 0.5 ev. In the region of 0.5 ev to 1.5 ev the effects of the 1.06-ev resonance of Pu^{240} were so strong that the Pu^{241} cross section could not be measured. The effects of possible nonuniformities of the sample have been considered, and it has been concluded that a maximum uncertainty of 5% in the cross section may exist as a result of nonuniformity. This maximum uncertainty occurs at the peak of the 0.264-ev resonance.

III. ANALYSIS

The fit to the Pu^{241} total cross section data is shown as the solid line in Fig. 1. In the application of the multilevel formula[7] to these data, the assumptions were made that in each of the two possible spin states formed by the addition of an s-wave neutron to Pu^{241}, there exists one neutron channel, two fission channels, and an essentially infinite number of radiative capture channels. Since only total cross section data were

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[7] C. W. Reich and M. S. Moore, Phys. Rev. **111**, 929 (1958).

[8] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).

[9] The isotopic analysis of the plutonium in the sample on October 14, 1959, expressed in atom percent, was as follows: Pu^{239}, 7.74±0.06; Pu^{240}, 10.42±0.10; Pu^{241}, 81.31±0.08; Pu^{242}, 0.53±0.05. The above errors, quoted at a 95% confidence level, were supplied by the Isotope Division of the Oak Ridge National Laboratory.

[10] O. D. Simpson, M. S. Moore, and F. B. Simpson, Nuclear Sci. and Eng. **7**, 187 (1960).

[11] D. J. Hughes and R. B. Schwartz, *Neutron Cross Sections, Brookhaven National Laboratory Report* BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

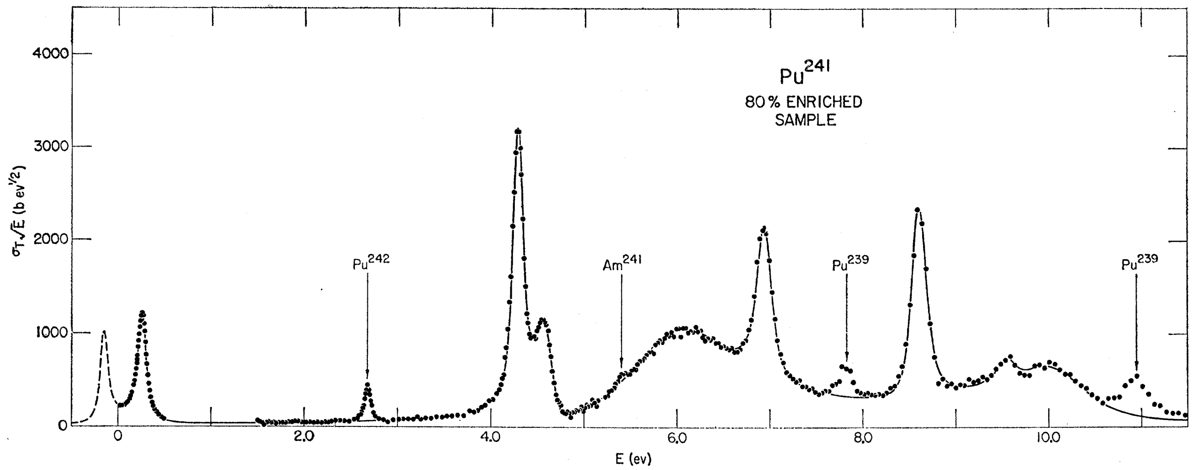


FIG. 1. The total cross section of Pu^{241}, multiplied by the square root of the neutron energy, as a function of neutron energy below 12 ev. The solid line shows the multilevel fit to the data. Doppler and resolution broadening corrections have been applied to the fit. The dashed line below zero neutron energy shows the negative energy level which was postulated in order to give a fit to the data below 0.5 ev. Resonances which are due to known contaminants are indicated. The strong resonance in Pu^{240} at 1.06 ev precluded the measurement of the Pu^{241} cross section in this region.

available for analysis, it was not possible to detect variations from resonance to resonance in the radiative capture widths. It was thus assumed that the radiative capture widths are constant, and equal to 0.040 ev. The parameters obtained from the multilevel analysis are shown in Table I.

The following procedure was used in obtaining the fit to the data:

(1) Trial parameters of E_λ, $\Gamma_{\lambda n}{}^0$, $\Gamma_{\lambda\gamma}$, $\Gamma_{\lambda f_1}$, and $\Gamma_{\lambda f_2}$ were selected, where E_λ is the characteristic energy of the resonance λ (not necessarily the energy at the maximum cross section), $\Gamma_{\lambda n}{}^0=\Gamma_{\lambda n}E^{-\frac{1}{2}}$ is the reduced neutron width, $\Gamma_{\lambda\gamma}$ is the radiative capture width (assumed to be 0.040 ev), and $\Gamma_{\lambda f_1}$ and $\Gamma_{\lambda f_2}$ are the fission widths in the two possible fission channels.

(2) A trial curve of σ_T as a function of energy was computed with the use of the multilevel formula.

(3) This theoretical curve $\sigma_T(E)$ was converted to velocity space, and Doppler broadening corrections were made by the numerical convolution technique described in Appendix A.

(4) The multilevel Doppler-corrected cross section curve was then converted to transmission as a function of time of flight, and instrumental resolution broadening corrections were made by a similar numerical convolution.

(5) The curve was then converted to $\sigma_T E^{\frac{1}{2}}$ as a function of energy, and compared to the experimental data shown in Fig. 1.

(6) The initial trial parameters were modified and the above steps were repeated as necessary until the solid curve shown in Fig. 1 was obtained.

The following features of the fit should be emphasized:

(1) The 0.264-ev resonance requires interference which is constructive below and destructive above the resonance energy. The energy variation of the interference required shows that it cannot be due to any

TABLE I. Multilevel parameters for the resonances in Pu^{241}. These parameters correspond to the fit to the data shown by the solid curve in Fig. 1. The quantity $\Gamma_{\lambda f_i}$ in the last column refers to either $\Gamma_{\lambda f_1}$ or $\Gamma_{\lambda f_2}$ as is appropriate. In the analysis, $\Gamma_{\lambda\gamma}$ is assumed to have the value 40×10^{-3} ev, and g was assumed to be $\frac{1}{2}$ for all resonances. Although a two-fission-channel, one-spin-state analysis was carried out, the results hold equally well for two spin states with a single fission channel in each. In this case, $\Gamma_{\lambda f_1}$ and $\Gamma_{\lambda f_2}$ are the fission widths in the first and second spin states, respectively, as discussed in the text. The relative signs of $(\Gamma_{\lambda n}\Gamma_{\lambda f_i})^{\frac{1}{2}}$ are those which correctly describe the interference in a single-fission-channel multilevel analysis, as described by Moore and Reich.[4]

Level (λ)	E_λ (ev)	$2g\Gamma_{\lambda n}{}^0$ (10^{-3} ev)	$\Gamma_{\lambda f_1}$ (10^{-3} ev)	$\Gamma_{\lambda f_2}$ (10^{-3} ev)	$\Gamma_{\lambda\gamma}$ (10^{-3} ev)	Relative signs of $(\Gamma_{\lambda n}\Gamma_{\lambda f_i})^{\frac{1}{2}}$
1	−0.160	0.0725	0	60	40	+
2	0.264	0.101	0	72	40	−
3	4.28	0.255	0	45	40	−
4	4.56	0.194	190	0	40	+
5	5.91	1.020	1350	0	40	+
6	6.94	0.218	0	95	40	+
7	8.60	0.268	0	70	40	+
8	9.56	0.035	0	100	40	−
9	10.20	0.400	1000	0	40	−

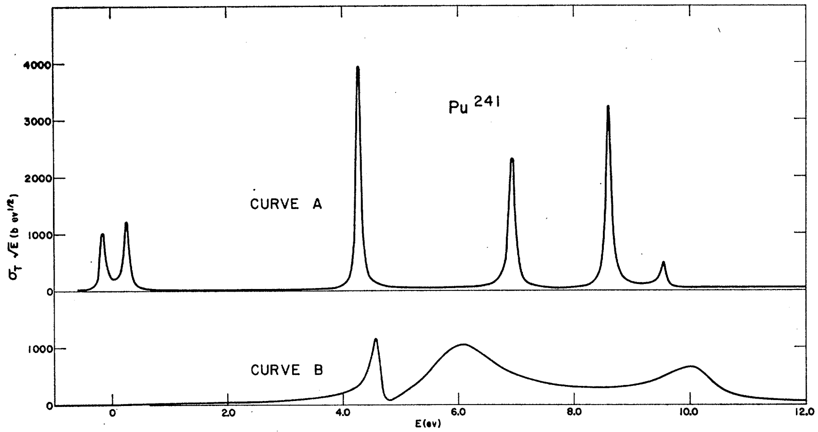


FIG. 2. The multilevel fit to the total cross section of Pu^{241}. Curve A shows those resonances which have been ascribed to one of the two possible spin states; curve B shows those which have been ascribed to the other. No Doppler- or resolution-broadening corrections have been applied to these curves.

positive energy resonance. The parameters of a negative energy resonance, shown in Fig. 1 as the dashed line, were selected to give the proper amount of interference and to give a value of 1020 barns for the fission cross section at 2200 m/sec.

(2) Although no experimental data were obtained between 0.5 and 1.5 ev, the calculated fit agrees well with the data below 0.5 and above 1.5 ev. It is therefore thought unlikely that any large resonance in Pu^{241} exists in this region.

(3) In the region of 4.8 ev, strong destructive interference is required, which has the effect of tilting the wide resonance near 6 ev. Complete interference is required between the 4.56 and the 5.91-ev resonance to describe these data.

(4) In the region of 8 ev, strong constructive interference is required, and only the wide resonances at 5.91 and 10.20 ev are of sufficient strength to provide this amount of interference.

(5) It is not possible to allow any of the narrow resonances between 6 and 10 ev to interfere to any large extent with the wide ones; any attempt to do so leads to the prediction of wild distortions which are not observed.

A further restriction was possible as a result of the above considerations. The parameters shown in Table I are those of a two-fission-channel analysis. However, the fit was achieved without mixing the channels, i.e., each resonance has a fission width which lies completely in one of the two possible channels. For no resonance is it required that the fission width have an amplitude in both channels. The above analysis has assumed for simplicity that all resonances belong to the same spin state and that two possible fission channels exist in this spin state. A more reasonable explanation is that the resonances belong to two spin states, each with but a single fission channel.[12] It may be pointed out, however, that the success in fitting the data with the assumption of a single fission channel does not necessarily limit the number to one, especially when so few levels are involved.

IV. DISCUSSION

The fit to the Pu^{241} cross section data in Fig. 1 is extremely good, but it is certainly not unique. Although the fission data[3] which have been obtained for Pu^{241} are not in disagreement with the above analysis, they do not permit detailed fitting over this energy region. Since only total cross section data were used in the present analysis, it is not surprising that a fit was achieved without requiring any variation in the radiative capture widths. Indeed, Pattenden and Harvey[13] have demonstrated that a reasonable fit to the total cross section of U^{233} can be obtained without varying $\Gamma_{\lambda\gamma}$ from level to level. But Moore and Reich[4] have found that if the fit is required to describe both the total and fission data for U^{233}, the radiative capture widths must be allowed to vary by more than a factor of two. For U^{235}, multilevel analyses of both total and fission data by Vogt[14] and by Shore and Sailor[5] also indicate a surprisingly large variation in the radiative capture widths. Even for Pu^{241}, where adequate fission data are available only over the 0.264-ev resonance,[3] a multilevel analysis of both the total and fission cross sections indicates that the radiative capture width is of the order of 0.032 ev and the fission width is 0.080 ev instead of 0.040 and 0.072 ev, respectively, as given in Table I. However, negligible changes are introduced into the fit to the total cross section data by this change in parameters.

A certain lack of uniqueness is also expected from the lack of knowledge of the spin states to which the resonances belong. The approach which was taken in the present analysis was guided somewhat by previous work on U^{233},[4] where the analysis also indicated that wide

[12] Although formally the two cases discussed are not the same, a comparison has been made of the multilevel calculations for the two cases which shows that they are experimentally indistinguishable for Pu^{241}.

[13] N. J. Pattenden and J. A. Harvey, *Proceedings of the International Conference on Nuclear Structure, Kingston, Canada* (University of Toronto Press, Toronto, Canada, 1960), p. 882.

[14] E. Vogt, Phys. Rev. **112**, 203 (1958); Phys. Rev. **118**, 724 (1960).

and narrow levels may belong to different spin states. The lack of uniqueness precludes the assignment of any error limits to the parameters.

In Fig. 2, the results of the analysis are displayed with the contributions from the two assumed spin states separated for clarity. No Doppler or resolution corrections have been applied to these curves. The lower curve shows the wide resonances, presumably of one spin state; the upper curve shows the narrow resonances of the other spin state. The wide resonances near 6 and 10 ev must interfere strongly and must of necessity be in the same spin state, in order to account for the large cross section at 8 ev. The narrow resonances between 6 and 10 ev cannot interfere strongly with the wide ones, so it seems reasonable to assume that they are of the other spin state. One of the two resonances of the doublet near 4.5 ev must interfere with the wide resonances. The present analysis assumes that it is the wider of the two, at 4.56 ev. The two resonances near zero neutron energy pose a somewhat different problem. They can be put in either spin state without affecting the fit. They must of course be in the same spin state, since they interfere with one another. It may be noted that fission fragment mass ratio studies on Pu^{241} by Regier *et al.*[15] are in agreement with this conclusion. It may also be of interest to note that the parameters of the negative energy resonance required to fit these data are in no sense anomalous, unlike those required for U^{235} and Pu^{239}.[14]

There are similarities in the cross sections of U^{233} and Pu^{241}, in that both exhibit wide and narrow resonances which appear to belong to different spin states. Both isotopes have spin $\frac{5}{2}$ and presumably positive parities, and the even-even compound nuclei U^{234} and Pu^{242} should thus have similar properties. According to the Bohr model of fission,[16] a compound nucleus which is undergoing fission is highly deformed. Much of its energy is potential energy of deformation, and the nucleus is "cold," i.e., not highly excited. The potential surfaces corresponding to low-lying states in the compound nucleus are expected to be preserved in the crossing of the fission barrier, and some information on the possible modes of oscillation of the highly deformed nucleus can be obtained from the energy levels observed near the ground state.

The spin states in the compound nuclei U^{234} and Pu^{242}, formed by the addition of an s-wave neutron to U^{233} or Pu^{241}, are 2^+ and 3^+. Since all the Pu^{242} resonances in this energy region exhibit a fission component,[3] it may be concluded that the fission threshold for both spin states in Pu^{242} is below the excitation afforded by $(Pu^{241}+n)$. Highly deformed even-even nuclei in this region of the periodic table have a very similar energy level structure near the ground state, and for purposes of this discussion, the energy levels of the better-known nucleus Pu^{238} are expected to be similar to U^{234} and Pu^{242}.[17] In Pu^{238}, three low-lying 2^+ states have been observed: at 44 kev, corresponding to the first rotational level of the ground state ($K=0$) configuration; at 985 kev, corresponding to the first rotational state of a β-vibrational ($K=0$) band; and at 1029 kev, corresponding to the first level of a γ-vibrational ($K=2$) band. A 3^+ level is observed at 1071 kev, and is identified as the second level of the γ-vibrational band. Neither the ground state nor the β-vibrational bands have a 3^+ level associated with them.

If, as the Bohr model suggests, the possible modes of oscillation do indeed correspond to the dispersion formalism concept of a fission channel, and if the excitation at the point of fission is such that only quadrupole vibration is significant, then the 2^+ spin state should fission through three channels and the 3^+ through one channel. In this case, following Wheeler,[18] the average fission width for the 2^+ spin state should be a factor of three larger than for the 3^+ spin state. In regions of large deformation, the ground state configuration might be expected to have a much lower fission threshold than the quadrupole vibrational bands, with a consequently much larger average fission width for the 2^+ state. In this case, a difference of a factor of considerably more than three would be expected in the average size of the fission widths belonging to the two spin states. The present analysis of Pu^{241} shows a factor of ten: for one spin state $\langle\Gamma_f\rangle$ is 0.847 ev; for the other spin state, $\langle\Gamma_f\rangle$ is 0.074 ev.

Since so few levels are represented in these averages, the size of the difference may not be significant. It is, however, significant that in Pu^{241} as well as in U^{233}, two types of resonances are observed, one type having large fission widths, the other having narrow widths, and that there is no appreciable interference between the two types.

APPENDIX A

It is very important that detailed Doppler and resolution broadening corrections be made to the calculated curve. For example, the corrected peak cross section of the 8.6-ev resonance was only $\frac{2}{3}$ of the uncorrected value. At any neutron velocity v_p the Doppler-corrected cross section, i.e., that which would be measured with perfect instrumental resolution, is related to the true cross section $\sigma_T(v)$ by the expression

$$\sigma_T(v_p)=(m/2\pi kT)^{\frac{1}{2}}\int_{-\infty}^{\infty}\sigma_T(v_p-v)\times\exp(-mv^2/2kT)dv, \quad (1)$$

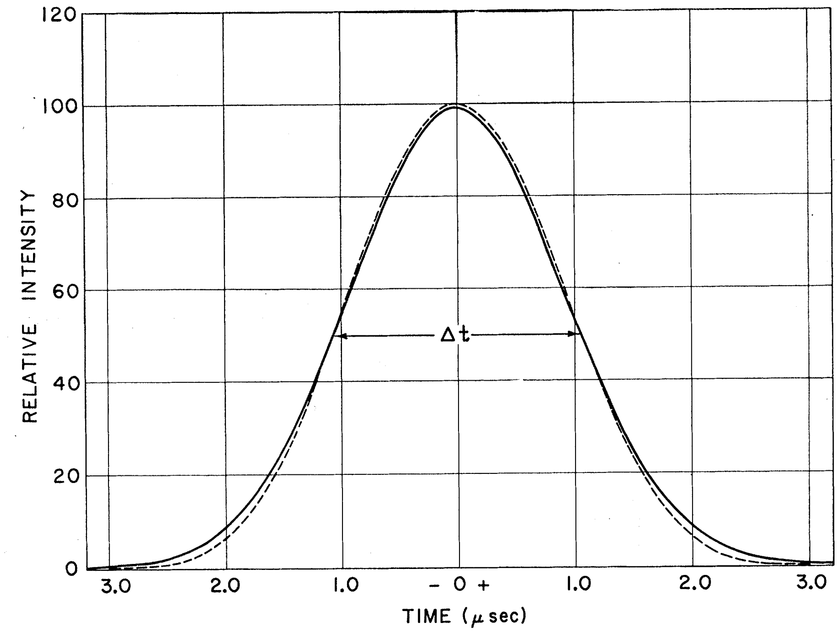
[15] R. B. Regier, W. H. Burgus, R. L. Tromp and B. H. Sorensen, Phys. Rev. **119**, 2017 (1960).

[16] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1956), Vol. 2, p. 151.

[17] R. K. Sheline, Revs. Modern Phys. **32**, 1 (1960).

[18] J. A. Wheeler, Physica **22**, 1103 (1956).

FIG. 3. Comparison of the fast-chopper resolution function with a Gaussian distribution. The solid curve shows the idealized fast-chopper resolution as a function of time. The dashed curve shows the Gaussian distribution which was used to represent the resolution function in making the necessary instrument resolution broadening corrections. The idealized chopper resolution function was obtained by the convolution of a symmetric triangle of appropriate width to represent the neutron burst, block functions to represent the channel widths and the counter thickness, and a Gaussian distribution to represent the uncertainty of detector pulse collection time. Both functions shown in this figure are normalized to unit area.



where m is the reduced mass of the neutron, k is the Boltzmann constant, and T is the effective temperature of the sample (see below).

In performing this convolution numerically, the integral was replaced by the summation

$$\sigma_T(v_p)=\sum_n \sigma_T(v_p-v_n)P(v_n), \tag{2}$$

where $P(v_n)$ is a step-function approximation to the normalized Gaussian function. For Doppler-broadening corrections, this function $P(v_n)$ is a constant (i.e., does not depend on v_p). It is thus convenient to perform the convolution in velocity space. This was done in effect by performing a four-point Lagrange interpolation of the calculated curve $\sigma_T(E)$ at energies E_n given by the expression

$$E_n=(E_0^{\frac{1}{2}}+nk)^2, \tag{3}$$

where E_0 is the initial energy under consideration, and k is a constant which is determined by the spacing of the points E_n and the width of the function $P(v_n)$.

Plutonium oxide has a Debye temperature of 403 ± 8°K[19] at room temperature. Correcting the ambient temperature T by the Debye temperature,[20] one calculates the effective temperature of the sample to be ≈ 375°K.

The correction of the calculated curve for instrumental resolution broadening was done by first converting the cross section to the transmission X by the expression

$$X(E_n)=e^{-N\sigma_T(E_n)}, \tag{4}$$

where N is the number of atoms/cm² for the sample which was used in the experimental measurement. A change of variable from energy to time of flight is convenient, since the measured transmission $X(t_p)$ at any flight time t_p is related to that which would be observed with perfect instrumental resolution by the expression

$$X(t_p)=\sum_{n'} X(t_p-t_{n'})R(t_{n'}), \tag{5}$$

where $R(t_{n'})$ is the observed instrumental resolution function, normalized to unit area. The function $R(t_{n'})$ is a convolution in time space of functions representing the channel width, fast chopper burst width, time jitter of the gating pulse, and collection time uncertainties in the detector system. The computed resolution function $R(t_{n'})$ for the operating conditions under which the data above 4 ev were collected is shown as the dashed curve in Fig. 3. The solid curve in Fig. 3 is a Gaussian function. Since the two curves agree very closely and since the idealized resolution function should include an additional small wing correction at these energies, a Gaussian representation was used as the function $R(t_{n'})$ in making the correction described.

The conversion of the calculated curve $X(E_n)$ given in Eq. 4 to time space is convenient since the function $R(t_{n'})$ is then approximately a constant. This was done by interpolating the curve $X(E_n)$ at energies $E_{n'}$ given by $E_{n'}=(E_0^{-\frac{1}{2}}-n'k')^{-2}$, where again E_0 is the initial energy and k' is a constant determined by the spacing of the points and the width of the curve $R(t_{n'})$.

[19] R. B. Roof, Jr., J. Nuclear Materials 2, 39 (1960).
[20] W. E. Lamb, Jr., Phys. Rev. 55, 190 (1939).

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Directional Correlation Measurements in Hf^{178}

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Measurements of the directional correlations of the 1335–93 kev, 1345–93 kev, and 1390–93 kev γ-γ cascades in Hf^{178} have been made, using a liquid source of 21-day W^{178} in equilibrium with 9.3-min Ta^{178}. It is shown that the data support the tentative spin assignments of Gallagher *et al.* The 1390-kev γ 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 γ 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 Ta^{178} isomer has recently been investigated by Gallagher, Nielsen, and Nielsen[1] (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 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 γ-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.

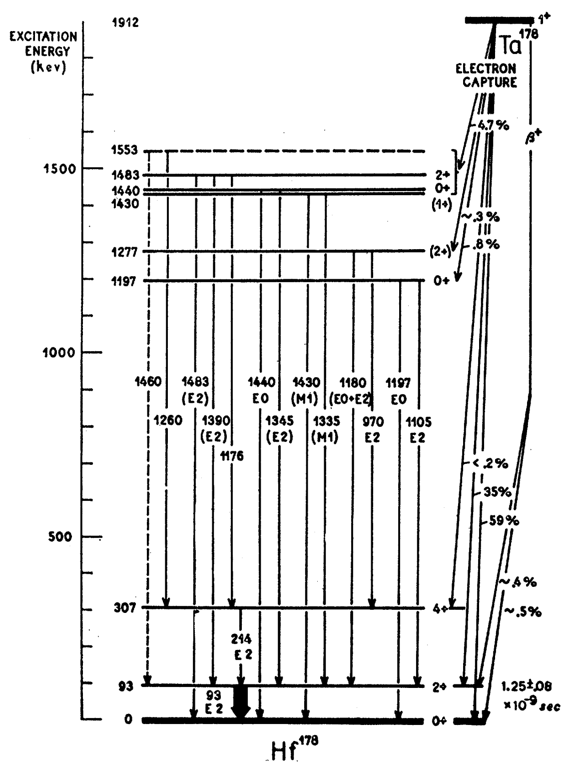

FIG. 1. Decay scheme of 9.3-min Ta^{178} according to GNN.

The purpose of this investigation is to obtain additional evidence on the spins of some of the high-lying levels of Hf^{178} by means of γ-γ directional correlation techniques and at the same time to collect as much information as possible about the multipolarities of the corresponding decay radiations. It is apparent from Fig. 1 that a number of γ-γ cascades are available in 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 γ 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 μC of 21-day W^{178} in equilibrium with 9.3-min 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 WF_8^{--} ions. The measurements were carried out using $1\frac{1}{2}\times1\frac{1}{2}$ in. NaI

[1] C. J. Gallagher, H. L. Nielsen, and O. B. Nielsen, Phys. Rev. 122, 1590 (1961).