the pertinent theoretical conversion coefficients computed by Hager and Seltzer.[8]

With the new value for α, the Mössbauer cross section for resonant absorption [Eq. (1)] is $\sigma_0=(1.40\pm0.05)\times10^{-18}$ cm^2, 6% higher than listed in the standard reference tables of Muir *et al.*[19] on the basis of earlier data.

[19] A. H. Muir, Jr., K. J. Ando, and H. M. Coogan, *Mössbauer Effect Data Index 1958–1965* (Interscience Publishers, Inc., New York, 1966), p. 126.

ACKNOWLEDGMENTS

The authors are indebted to Professor G. T. Emery of Indiana University for calling this problem to their attention. It is a pleasure to thank him and Dr. M. L. Perlman of Brookhaven National Laboratory for several very helpful discussions. H. R. Bowman of the Lawrence Radiation Laboratory, Berkeley, has been most generous with advice on the construction of the spectrometer, largely of his design.

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Nuclear Pairing Energy of Transition Nucleus Pu^{240}†

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Fission-fragment angular distributions are measured for the fission of Pu^{239} induced by monoenergetic neutrons of energies 150, 400, 475, 550, 600, 700, 800, 900, 1000, 1100, 1200, 1350, and 1500 keV (energy resolution, ±25 keV). Theoretical calculations are utilized to determine the parameter K_0^2 at each excitation energy from the observed angular distributions. The values of K_0^2 rise sharply from a value of 5–6, for neutron energies less than or equal to 600 keV, to an approximately constant value of 13, for neutron energies in the range 1.3–2.0 MeV. The break in the K_0^2 curve occurs at about 2.2 MeV above the fission threshold, and is interpreted as the beginning of two-quasiparticle excitations of the highly deformed transition nucleus Pu^{240}. This result suggests that the pairing energy gap is increased for large nuclear deformations.

I. INTRODUCTION

THE capture of a neutron in Pu^{239} produces an even-even fissioning nucleus. It is well known that Pu^{239} has a large thermal-neutron fission cross section. Studies with resonance neutrons have revealed two groups of fission resonances.[1] The fission widths of each group of resonances is reasonably well fitted with a Porter-Thomas distribution, indicating a single fission channel for each spin state.[1] Furthermore, from the measured average fission width, one deduces that the 0+ channel is completely open, whereas the 1+ channel is only partially open. The 0+ level is thought to define the fission threshold for an even-even transition nucleus. The above information suggests that thermal and resonance neutrons have insufficient energy to excite quasiparticle excitations in the transition nucleus Pu^{240}.

† Work supported by the U. S. Atomic Energy Commission.

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The present experiments were initiated to look for the two-quasiparticle threshold in the intermediate transition nucleus. Measurements of fission-fragment angular distributions as a function of energy reveal information on the transition states of a nucleus as it passes over the fission barrier.[2–6] These transition states

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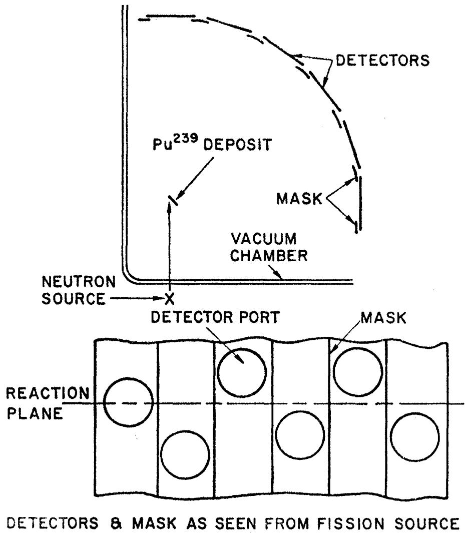

FIG. 1. Schematic diagram of experimental arrangement.

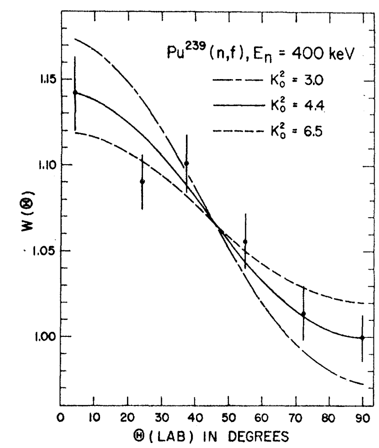

FIG. 2. Experimental and theoretical fission-fragment angular distributions for fission of Pu^{239} induced with 400 ± 25 keV neutrons. The solid circles are experimental points, and the lines correspond to theoretical results with different values of K_0^2. Perey-Buck neutron transmission coefficients (Ref. 12) are used in these theoretical calculations.

consist of collective and particle excitations of the fissioning nucleus at the so-called saddle-point deformation. In the present experiments, we are especially interested in the first few MeV of excitation energy above the fission threshold. In this energy region, the effects of superfluidity due to nuclear pairing are expected to be most important.

Fission-fragment angular distribution measurements are a sensitive probe of the threshold energy for two-quasiparticle excitations. In the energy region of collective excitations, K_0^2 is expected to be small. However, a break in K_0^2 is expected to occur at an energy corresponding to the onset of two-quasiparticle excitations. The fission-fragment angular distributions for the Pu^{239} (n,f) reaction are rather insensitive to competition from neutron and γ-ray emission and permit an unambiguous determination of the excitation energy dependence of the transition spectrum.

The fission-fragment angular distribution is uniquely determined by the quantum numbers of the transition state allowing for two assumptions.[2–6] First, the fission fragments separate along the extended nuclear symmetry axis; and, second, K is a good quantum number in going from the transition nucleus to scission. The probability distribution in direction is then equal to the probability distribution in direction of the nuclear-symmetry axis, which is characterized by I (total angular momentum), K (projection of I on the nuclear-symmetry axis) and M (projection of I on a space-fixed axis, which in our case is the neutron beam direction).

In all of our calculations, we have assumed the K distribution to have a Gaussian form characterized by the parameter K_0^2:

$$F(K) \propto \exp(-K^2/2K_0^2). \tag{1}$$

Detailed analysis of the energy dependence of the fission-fragment anisotropy from the Pu^{239} (d,pf) reaction[7] has given evidence of structure in a plot of K_0^2 as a function of excitation energy for the transition nucleus Pu^{240}. From the energy position of one of the breaks in the K_0^2 curve, the magnitude of the pairing gap 2Δ in the highly deformed transition nucleus Pu^{240} was estimated to be 2.6 MeV.[7] However, some questions have been raised about the interpretation of the various breaks in the energy-dependent K_0^2 curve.[6,8] In order to eliminate the possibility that some of the structure in K_0^2 is associated with the (d,p) reaction mechanism, we have reinvestigated the pairing energy of the Pu^{240} transition nucleus by a study of the Pu^{239} (n,f) reaction. Because of the important implications of a deformation-dependent pairing energy on nuclear structure in general, one wishes to obtain supporting evidence from at least two different types of reactions. Some information on fission-fragment angular distributions from the Pu^{239} (n,f) reaction has been reported previously.[9,10] However, the low-energy measurements are either missing[9] or in disagreement[10] with our results.

II. EXPERIMENTAL PROCEDURE

A schematic drawing of the experimental arrangement is shown in Fig. 1. Monoenergetic neutron beams were produced from the $Li^7(p,n)Be^7$ reaction. Protons were accelerated in the Argonne Reactor-Physics Van de Graaff and bombarded a lithium target prepared by

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TABLE I. Fission-fragment angular distributions for the $Pu^{239}(n,f)$ reaction at several neutron energies.

	$W(\theta)$ Experimental					
E_n (keV)	3.7°	24.0°	37.2°	54.7°	72.3°	90.0°
150±25	1.066±0.014	1.041±0.014	1.056±0.015	1.001±0.014	0.990±0.014	1.000±0.012
400±25	1.142±0.021	1.090±0.016	1.101±0.017	1.056±0.016	1.014±0.016	1.000±0.013
475±25	1.108±0.012	1.090±0.013	1.058±0.012	1.009±0.012	1.040±0.013	1.000±0.010
550±25	1.128±0.013	1.116±0.014	1.090±0.014	1.019±0.013	1.019±0.013	1.000±0.011
600±25	1.148±0.011	1.112±0.012	1.083±0.011	1.052±0.011	1.033±0.012	1.000±0.011
700±25	1.113±0.014	1.080±0.015	1.048±0.015	1.014±0.014	1.000±0.015	1.000±0.012
800±25	1.132±0.012	1.104±0.012	1.079±0.012	1.035±0.011	1.033±0.012	1.000±0.010
900±25	1.111±0.012	1.101±0.012	1.071±0.012	1.057±0.012	1.002±0.012	1.000±0.010
1000±25	1.123±0.015	1.099±0.015	1.082±0.015	1.028±0.014	1.002±0.015	1.000±0.012
1100±25	1.128±0.012	1.105±0.012	1.101±0.012	1.027±0.011	1.037±0.012	1.000±0.010
1200±25	1.122±0.013	1.104±0.014	1.066±0.014	1.027±0.013	0.993±0.014	1.000±0.011
1350±25	1.124±0.013	1.109±0.013	1.073±0.013	1.025±0.012	1.024±0.012	1.000±0.010
1500±25	1.153±0.013	1.147±0.013	1.121±0.013	1.046±0.012	1.000±0.012	1.000±0.010

vacuum evaporation. The neutron energy spread, determined mainly by the lithium target thickness, was ±25 keV. The neutrons impinged on a Pu^{239} target (99.7% isotopically pure) prepared by a painting technique on a 0.001-in. aluminum backing foil. The Pu^{239} target has a thickness of 0.5 mg/cm², and the diameter was restricted to $\frac{1}{8}$ in. by a mask.

The fission fragments from the Pu^{239} (n,f) reaction were detected at six angles, simultaneously with surface barrier solid-state detectors (see Fig. 1). The six detectors were each centered in the scattering chamber at one of the following angles θ to the neutron-beam direction, 3.7°, 24.0°, 37.2°, 54.7°, 72.3°, and 90.0°, respectively. The detectors, approximately 1.57 cm in diameter, were located 7.30 cm from the Pu^{239} target.

The collimating system restricted the image of the proton beam spot on the lithium target to a diameter less than $\frac{1}{8}$ in. With the angular spread in the neutron beam direction and the above detector arrangement, the over-all angular resolution was approximately 8° full width at half-maximum.

III. EXPERIMENTAL RESULTS

The fission-fragment angular distributions for the Pu^{239} (n,f) reaction were measured at average neutron-irradiation energies of 150, 400, 475, 550, 600, 700, 800, 900, 1000, 1100, 1200, 1350, and 1500 keV. The beam energy spread was ±25 keV. The data for each were gathered in a series of runs in order to check the over-all consistency of the data at each energy. A summary of the results is given in Table I. For three energies, 400, 900, and 1350 keV, the angular distributions are plotted in Figs. 2, 3, and 4, respectively.

After mounting the Pu^{239} target in the vacuum chamber, the relative areas of the six detectors were determined by irradiation with thermal neutrons. With s-wave neutron capture, the angular distribution is isotropic. Since the relative detector areas were determined with the target in an identical geometrical position as that of the various runs, all corrections due to possible irregularities in target thickness, exact positioning of target, etc., cancel.

Since the fission cross section of Pu^{239} for thermal neutrons is much larger than it is for 100–1500-keV neutrons, the possibility of contamination by thermal-neutron-induced fission was investigated. The effect of such a contamination is to lower the anisotropy and to give values of K_0^2 that are too large. However, at selected neutron energies, the fission rate was measured with the detector chamber alternately bare and covered

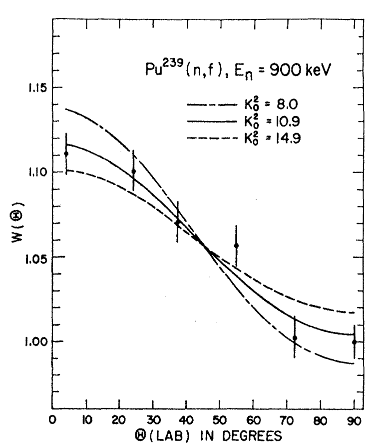

FIG. 3. Same as caption of Fig. 2 except 900±25 keV neutrons.

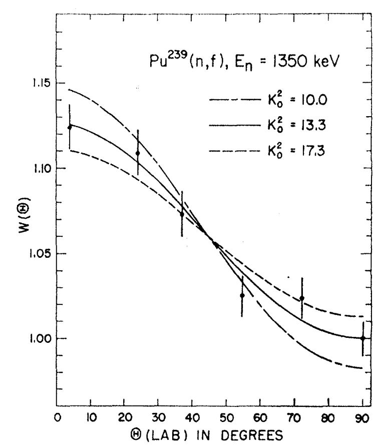

FIG. 4. Same as caption of Fig. 2 except 1350±25 keV neutrons.

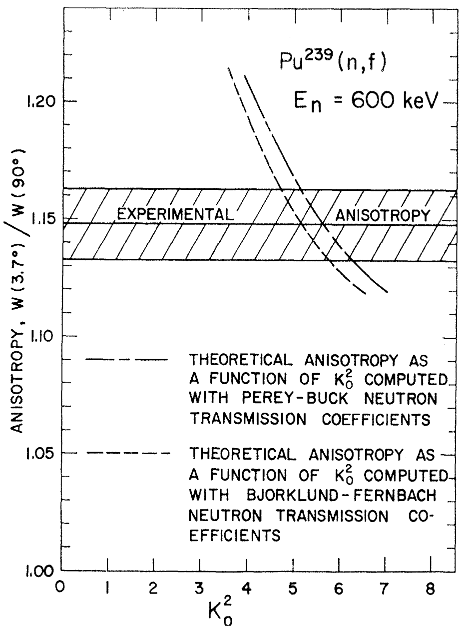

FIG. 5. Experimental and theoretical fission-fragment anisotropies $W(3.7°)/W(90°)$ for fission of Pu^{239} induced with 600 ± 25 keV neutrons. The theoretical anisotropies as a function of K_0^2 are calculated with Perey-Buck and Bjorklund-Fernbach neutron transmission coefficients (Ref. 12).

with 0.020 in. of cadmium. An upper limit of 5% of the fission events is due to thermal-neutron fission. The fraction of fission events induced by neutrons scattered by the air, walls, support structure, etc., was determined by measuring the dependence of the fission rate on distance from the neutron source. The results indicate that essentially all the neutrons came directly from the source with a probable error of 5%.

The fraction of fission events produced by neutrons from the $Li^7(p,n)Be^{7*}$ reaction, which leaves Be^7 in its first excited state is zero for $E_n \leq 500$ keV. At the higher energies, the fraction is not negligible; however, the anisotropy is quite insensitive to neutron energy; and, therefore, we have neglected to make a correction for the second neutron group.

IV. THEORY

The fissioning-transition nucleus is completely characterized by the quantum numbers I (total angular momentum), M (projection of I on a space-fixed axis to be designated as the neutron-beam direction), and K (projection of I on the nuclear-symmetry axis). If it is assumed that the fragments separate along the nuclear symmetry axis, the angular distribution is uniquely determined by the above quantum numbers.[2–6]

If a compound state (I,M) fissions through a transition state K, the angular distribution is given by the square of the rotational part of the collective wave function:

$$W_{M,K}{}^I(\theta)=\frac{(2I+1)}{4\pi}|d_{M,K}{}^I(\theta)|^2. \tag{2}$$

The $d_{M,K}{}^I(\theta)$ functions are given by[5]

$$d_{M,K}{}^I(\theta)=\{(I+M)!(I-M)!(I+K)!(I-K)!\}^{1/2}\times\sum_X\frac{(-1)^X(\sin\frac{1}{2}\theta)^{K-M+2X}(\cos\frac{1}{2}\theta)^{2I-K+M-2X}}{(I-K-X)!(I+M-X)!(X+K-M)!X!}, \tag{3}$$

where the sum is over $X=0, 1, 2, 3 \cdots$ and contains all terms in which no negative value appears in the denominator of the sum for any one of the quantities in parentheses. Therefore, the fission-fragment angular distribution offers a direct source of information on the spectrum of quantum states associated with the transition nucleus.

The relative fission-fragment angular cross section is given by[11]

$$W(\theta)\alpha\sum_{JIM}\sum\sum P(J;IM)W(I,M;\theta). \tag{4}$$

The first term is a partial probability of formation of a state (I,M) of the compound nucleus from a particular J value:

$$P(J;IM)=\frac{(2J+1)\{T_{l=J-1/2}(E)+T_{l=J+1/2}(E)\}\sum_\mu\sum_\sigma|C_{\sigma,\mu,M}{}^{J,I_0,I}|^2}{(2s+1)(2I_0+1)\sum_J(2J+1)\{T_{l=J-1/2}(E)+T_{l=J+1/2}(E)\}}. \tag{5}$$

The transmission coefficients $T_l{}^J(E)$ are derived from the optical model with spin-orbit interaction, where $\mathbf{J}=\mathbf{l}+\mathbf{s}$ and s is the neutron spin with projection σ on the space-fixed axis. The target spin and its projection on the space-fixed axis are denoted by I_0 and μ, respectively. The total angular momentum I of the compound nucleus is given by $\mathbf{I}=\mathbf{J}+\mathbf{I}_0$, and the projection of I on the space-fixed axis is given by M. The quantity $C_{\sigma,\mu,M}{}^{J,I_0,I}$ is a Clebsch-Gordan coefficient. The second term $W(I,M;\theta)$ gives the angular distribution of the fission fragments emitted from the compound state (I,M):

$$W(I,M;\theta)=\sum_{K=-I}^{+I}\exp(-K^2/2K_0^2)(2I+1)|d_{M,K}{}^I(\theta)|^2\Big/\sum_{K=-I}^{+I}\exp(-K^2/2K_0^2). \tag{6}$$

The relative cross section at angle θ is obtained by summing the product $P(J;IM)\ W(I,M;\theta)$ over all values of M, I, and J.

V. RESULTS OF CALCULATIONS AND DISCUSSION

Computer calculations were utilized to search for a value of K_0^2 in Eqs. (4)–(6) that minimized the difference between the theoretical and experimental fission-fragment angular cross sections (χ^2 criterion). Examples of comparisons between theory and experiment are shown for neutron energies of 400, 900, and 1350 keV in Figs. 2–4, respectively. On each figure, the best fit is shown (derived K_0^2) as well as the angular distributions for values of K_0^2 smaller and larger than the derived value of K_0^2. In Fig. 5, we show curves of the calculated anisotropy ratio $W(3.7°)/W(90°)$ as a function of K_0^2 for $E_n=600$ keV and two sets of neutron transmission coefficients. The experimental anisotropy is also shown in this figure. From the experimental error, one can evaluate the uncertainty in the derived value of K_0^2.

Values of K_0^2 derived at neutron energies of 100, 400, 475, 550, 600, 700, 800, 900, 1000, 1100, 1200, 1350, and 1500 keV from the above type calculation are plotted as solid circles in Fig. 6 as a function of neutron energy. Values of K_0^2 are also given for neutron energies of 1750, 2000, 2250, 2500, 2750, and 3000 keV as open circles. These values were derived from Simmon's anisotropy data[9] in the following way. At neutron energies 1.00 and 1.50 MeV, our anisotropy ratios $W(3.7°)/W(90°)$ are 1.050 times larger than Simmon's anisotropy ratios $W(10°)/W(90°)$. We assume this difference results from an incorrect calibration of the detector sizes at the two angles in Simmons's experiment. We, therefore, multiply Simmons's anisotropy data by the above normalization factor to give the anisotropy ratios $W(3.7°)/W(90°)$, which were used for the computation of K_0^2 (open circles on Fig. 6).

All of the results shown in Fig. 6 were computed with Perey-Buck neutron transmission coefficients[12] (with spin-orbital potential). Calculations were performed also with Bjorklund-Fernbach neutron transmission coefficients.[12] The general results are similar to the results shown in Fig. 6 except that the value of K_0^2 at each energy is decreased by about $\frac{1}{2}$ unit (see Fig. 5).

The values of K_0^2 are of the order of 5 to 6 for neutron energies up to 600 keV. Since the fission threshold of Pu^{240} is 4.90 MeV[13] and the neutron binding energy is 6.47 MeV,[14] the excitation energy in the transition nucleus for 600-keV neutrons is 2.17 MeV. We interpret the small K_0^2 values at excitation energies less

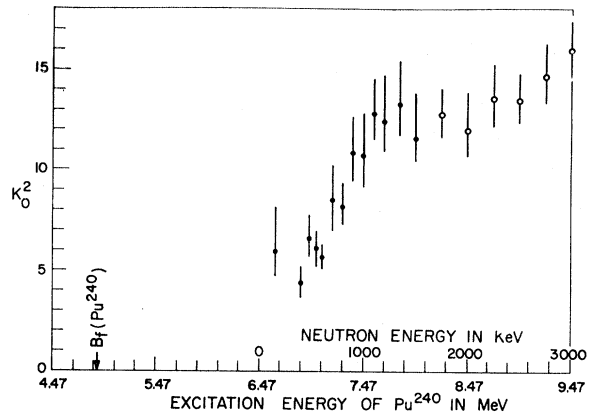

FIG. 6. The derived values of K_0^2 are plotted both as a function of neutron energy in keV and as a function of the excitation energy of Pu^{240} in MeV. The solid circles are derived from the present experiments, and the open circles are derived from data of Simmons (Ref. 9). The data of Simmons have been normalized to our data as described in the text.

than 2.2 MeV to mean that only collective levels are excited in this energy interval. Although the ground-state band of an even-even transition nucleus has $K=0$, at excitation energies of 1.7 to 2.2 MeV, a number of collective vibrational excitations and their rotational bands are expected to contribute. Our value of $K_0^2=5$ to 6 in this energy interval is in reasonable agreement with values from the Pu^{239} (d,pf) reaction[7] in the same energy region.

The value of K_0^2 increases rapidly from 600 to 1100 keV and reaches a plateau value of about 13 at 1100 keV. In the $N=5$ oscillator shell, one has single-particle states $1h_{11/2}$, $1h_{9/2}$, $2f_{7/2}$, $2f_{5/2}$, $3p_{3/2}$, and $3p_{1/2}$.[15] In a deformed nucleus where K is a good quantum number, the average value of $\langle K^2\rangle$ in the 5th oscillator shell is readily computed for the above states to be 7 per quasiparticle. A similar calculation for the $N=6$ oscillator shell gives $\langle K^2\rangle=9$ per quasiparticle. Hence, the experimental value of $K_0^2=13$ is consistent with an average two-quasiparticle excitation in the 5th oscillator shell. If the excitation energy is sufficient to break either a neutron or proton pair, $\langle K^2\rangle$ is a weighed average over two neutron and two proton quasiparticles.

The values of K_0^2 begin to increase markedly at an excitation energy of 2.2 MeV in the Pu^{240} transition nucleus (this energy is based on $B_f=4.90$ MeV). We assign this energy of 2.2 MeV as the threshold energy for two-quasiparticle excitations in the Pu^{240} transition nucleus. The above value of the two-quasiparticle threshold determined from the Pu^{239} (n,f) reaction is in good agreement with a recent value[16] of the same quantity from the Pu^{239} (d,pf) reaction. In Fig. 6, one expects a second rise in K_0^2 at the four-quasiparticle

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threshold. Although not shown in this figure, the angular distribution data of Simmons[9] do indicate that K_0^2 rises again beginning with 3.0-MeV neutrons. The data represented by circles in Fig. 6, however, must be treated with some caution since a normalization (described earlier) is involved.

The pairing energy of nuclei decreases with increasing A.[17] It has been suggested[18,19] that the pairing effect is associated with a matrix element predominately in the nuclear surface. With increasing A, the surface-to-volume ratio ($\alpha A^{-1/3}$) decreases; and, hence, the pairing energy decreases as one goes from light to heavy nuclei.[18] This suggests that the pairing gap should increase for a particular nucleus as one goes from the equilibrium deformation to the much larger nuclear deformation of the transition nucleus.[19] The present experiments support an enhanced pairing gap in the more deformed Pu^{240} transition nucleus. The experimental magnitude of the enhancement is consistent with a recent calculation[20] which assumes that the pairing force strength in nuclei varies proportionally to the area of the nuclear surface. If the larger pairing energy is due to the influence of the nuclear surface, one expects even more pronounced differences in the pairing gap between equilibrium and transition nuclei in the vicinity of lead. These transition nuclei are deformed essentially to the scission configuration with very large surface-to-volume ratios. Preliminary results[21] in this region of the periodic table do indicate a large pairing gap consistent with the enhanced pairing energy for very deformed heavy nuclei presented here.

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Intensities and Angular Distributions of Ground-State Rotational Transitions Excited in (α,3n) and (α,4n) Reactions*

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Relative intensities measured for rotational transitions in the ground-state bands are reported for residual even-even rare-earth nuclei excited in (α,3n) and (α,4n) reactions. The incident α-particle energy was typically ~40.5 MeV, and the targets were isotopes of Gd, Dy, Er, and Yb. The angular distributions of the rotational radiations were found to be in accord with theoretical expectations. The relative intensities of the lines were, however, inconsistent with the implications of simple statistical models. More angular momentum is apparently carried away by photon transitions connecting higher-lying states than one might expect. Possible reasons for this discrepancy are briefly discussed.

I. INTRODUCTION

It has been known for some time that ground-state rotational bands of even-even distorted nuclei can be easily excited in bombardments involving the deposition of considerable angular momentum.[1–4] The purpose of the present measurements was to use the relative intensities of the lines excited in such bands to show how much of the initial angular momentum in the system remains at the end of the de-excitation, i.e., at the point of entry into the ground-state band of the final residual nucleus.[5] Such measurements therefore complement those of angular distributions of evaporated particles[6,7] and photons[8] which also give some information about the rate of loss of angular momentum during nuclear de-excitation. In the past, the relative intensities of low-lying isomers have sometimes been used as

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