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INTERMEDIATE STRUCTURES IN FISSION -
SPECTROSCOPY OF SHAPE ISOMERS

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Abstract - The spectroscopy of rotational and intrinsic states built on the spontaneously fissioning shape isomers is reviewed on the basis of available experiments. For doubly even nuclei, the "ground" state rotational band, K-isomers, vibrational excitations and compound levels are discussed. For odd-neutron nuclei the single-neutron excitations are discussed and compared to the Nilsson diagram at large distortions (e = 0.6).

I - INTRODUCTION

This paper is a summary of what we know about the spectroscopy of the shape isomers in the uranium region.

For many years we have thought that if you want to induce fission in a heavy nucleus, you have to pull it out. The surface tension which may include shell effects resists this, but Coulomb repulsion helps. When you pass a critical elongation, the saddle, the nucleus spontaneously flies apart in two pieces. Many nuclei behave that way. However, starting with the pioneering work of today's chairman and his co-workers, we have learned that some nuclei, from thorium to berkelium, will resist as usual when you pull them, they will reach a critical elongation, but then they will go - snap - into a new quasistable configuration with a ratio of axes of about 1:2, and you have to pull once more to induce fission. This second shape is not all that stable but apart from that, it is as good a nucleus as any ground state we know of. In a sense it is a better nucleus. This is illustrated in

FIG.1 - The spontaneously-fissioning shape isomers have higher symmetry than ordinary non-spherical nuclei. This should be reflected in their excitation spectra.

figure 1: Since the days of antiquity it has generally been agreed that the sphere, which has the highest symmetry, is the most beautiful shape. If the neutron and proton numbers are right,
shell effects associated with the symmetry, will ensure that the nucleus takes the most beautiful form, the sphere. Conversely, if the nucleon numbers are wrong, the same shell effects will force the nucleus to avoid the sphere and assume a wrong ground state shape with lower symmetry. The shape isomers occur in nuclei with nucleon numbers wrong with respect to the sphere. They have found a shape with a symmetry - a ratio of axes of 1:2 - for which their nucleon numbers are right. In this sense they are more beautiful than the ordinary non-spherical ground states.

The following is a status report of the spectroscopy of this isomeric shape. It is still in an embryonic stage, but it may be expected that this spectroscopy will become something more than a trivial extension of what has been learned already from the spectra of ordinary deformed nuclei. The higher symmetry - the "magic" ratio of axes - should influence the spectra and lead to novel features which it will be of special interest to explore.

II - SPECTROSCOPY OF DOUBLY EVEN SHAPE ISOMERS

Plutonium 240 is the best studied case. In a measurement, which I will not hesitate to call the experiment of the year, Specht, Konecny, Heunemann and Weber have identified the E2 conversion lines of the ground state rotational band, built on the shape isomeric state. The isomer is formed in the $^{238}$U(a,2n) reaction and the traditional technique of measuring the low lying transitions, forming the "heavy traffic line" towards the ground state, is used. Figure 2 illustrates the situation. Below, the fraction of the total cross section that goes through various channels is given. In a typical example, $^{172}$Yb, it is 90 per cent. For the $^{240}$Pu isomer it is $10^{-2}$ per cent, so the traffic is not really heavy at all. How the Munich group solved this difficulty is described in ref.[1]. Their result is given in figure 3. The band has a moment of inertia more than twice larger than the corresponding band in the first well and the energy spacings are extremely close to the adiabatic, I(I+1), limit.

Figure 4 illustrates another important spectroscopic measurement, due to Limkilde and Sletten [2]. In the first well of $^{238}$Pu, as in

![Flow diagram for the deexcitation of a fissile compound nucleus with two competing wells.](image)
Isomerism due to the K-selection rule is a well established phenomenon in non-spherical nuclei (first well). Such isomers should also and are indeed found in the second well. (The $K,I^+ = 4.4$ isomer in the first well $^{238}_{\text{Pu}}$ was identified by Bengtson et al. [3].)

For many other doubly even nuclei, there exists an isomeric state which is generated by breaking a neutron pair and coupling the two spin projections to a large K-value which hinders the $\gamma$-decay to the ground state levels. The excitation energy is that required for breaking a pair. It measures the magnitude of the neutron energy gap $2\Delta_n$ with a determinacy of about 0.2 MeV. In $^{238}_{\text{Pu}}$ there are also two fission isomers. Although the $K$-values remain unknown, the authors of ref. [2] have successfully measured the energy difference to be 1.320.3 MeV. Figure 5 compares their result to the situation in the first well for various $K$-isomers of doubly even nuclides in the vicinity of $^{238}_{\text{Pu}}$. The figure also shows the odd-even mass difference $P_N$ derived from the neutron separation energies, another experimental measure of the neutron energy gap. (The proton gap is nearly always larger.)

By ingenious extension of traditional techniques, we thus know the ground state rotational band and the (neutron) energy gap for the shape isomer. The specific nature of the isomeric state, enclosed as it is between two relatively low barriers, provides a unique possibility of measuring the spectrum of vibrational states at higher energies - in the energy interval from about one to zero MeV below the barrier top. The state must be predominantly of the $\beta$-vibrational type, i.e. of the stretching type that leads to fission. Figure 6 shows a textbook version of a double humped barrier [7]. It is symmetric and only one degree-of-freedom is considered. The well between the barriers holds a number of quasibound states. They can decay by tunnelling through the barrier and hence have finite widths which increase as the barrier top is approached. Waves coming from the left will penetrate the double barrier, and if the energy of the incoming wave matches the energy of the bound state, the penetrability will be unity, as shown to the right in figure 6. An asymmetric barrier, figure 7, which is more realistic, will also give resonance transmission, but with peak transmissions lower than unity. As before, the energy width of the transmitted wave is equal to the width of the quasibound state and hence to the inverse lifetime. Compound motion in the first well (not shown) simulates a broad-band spectrum of incoming waves. If the compound levels in the first well are populated uniformly, by a $(d,p)$ process for example, one will observe an enhanced fission probability at the resonance.
energies. Figure 8 shows such resonances in $^{240}$Pu, ref.[8]. They are rather broad. Therefore one would at first be inclined to say that the barriers defining the resonances must be quite thin. This actually is not so. There exists a measurement [9] of the 4.9 MeV resonance made with higher resolution, figure 9. What appears to be a broad Lorentzian shape has indeed fine structure with energy spacings comparable to the total level spacings expected in the second well at this excitation energy: 2.9 MeV. So, we are reminded of the oversimplification introduced when treating the problem in one degree-of-freedom only. It is no real surprise that the $\beta$-vibrational motion couples to other degrees-of-freedom and leads to configuration mixing. The measurement, figure 9, shows how much mixing there is, and that is a piece of information which it has never been possible to extract from measurements of vibrations of the ordinary shapes, belonging to the first well. In fact, two-phonon or higher vibrational states of deformed nuclei have not yet been identified in ordinary deformed nuclei. The specific property of the stretching vibrations in the second well, namely that they and only they lead to fission, makes it possible to study them separately. In addition, a measurement of the fragment angular distributions should in principle lead to the determination of the quantum numbers, $I,K$ of the vibrational states. In practice this has only been possible to a limited degree. Here, $\gamma$-ray induced fission with monochromatic $\gamma$-rays has the advantage of picking out the $1^-$ and perhaps $2^+$ resonances exclusively.
FIG. 8 - Resonance barrier penetration in $^{242}$Pu. Compound levels in the first well are populated in the (d,p) process after which fission takes place in competition with γ-emission. The total proton spectrum is shown above, the fraction of protons followed by fission is the lowest curve, ref. [8].

There is some interesting recent work by Knowles along this line [10].

At still higher energies the coupling of the fission motion to other degrees-of-freedom is expected to wash out the vibrational resonance structure completely. This is equivalent to compound nucleus motion. In the neutron induced fission of $^{239}$Pu with resonance neutrons, one observes intermediate structure in the fission probability of the $1^+$ capture states [11,12]. The spacing, 0.46 keV, reflects the level density of the $1^+$ compound states in the second well. Figure 10 summarizes the spectroscopic information obtained so far for a typical doubly even shape isomer, $^{240}$Pu. (The K-isomer actually belongs to $^{238}$Pu, but this is no serious distortion of facts.)

Compared to the wealth of spectroscopic information existing on the excitations of the ordinary ground state shape, the results are modest. But it is a beginning. With respect to the higher lying vibrations, the picture is actually quite advanced, as stressed above.

We conclude this section on doubly even nuclei with a comparison of experiment and a recent theoretical calculation due to A. Sobczewski [13], figure 11.

FIG. 9 - High resolution measurement of the 4.9 MeV resonance shown in figure 8, ref. [9].
Above, the experimental moments of inertia are compared to a cranking calculation, including pairing. The deformation $\varepsilon$ of the isomer is assumed equal to 0.6 on theoretical grounds. That is where the second minimum lies according to the Strutinsky type calculations [14,15,16]. It corresponds to a ratio of axes of 1:2. The agreement is good, but it is not possible to say whether a pairing strength proportional to the surface area, $G \propto S$, should be preferred over a constant pairing strength, $G \propto const$. Below, on figure 11, the calculated neutron energy gap in the two versions, $G \propto S$ and $G \propto const.$, is compared to the experimental values of the (neutron) energy gap. (Once more, a possible difference between $^{238}$Pu and $^{240}$Pu is assumed to be negligible.) The agreement is satisfactory, but uncertainties are again too large to allow for a definite choice between the two versions of the pairing strength.

A comparison of the experimental quadrupole moment — including its sign — and hence of the deformation of the shape isomer with the theoretical value would have been most interesting. This regrettably has to await further progress in experimental techniques.

III - SPECTROSCOPY OF ODD-A SHAPE ISOMERS

The odd-A nuclei can be thought of as a doubly even core to which an extra particle is coupled. With the core in its ground state the odd particle determines the spin, $\Omega$, value and parity of the system as a whole. Different single-particle configurations, each with a given $\Omega$ value, carry a different energy which may vary a good deal with deformation. This is the specialization energy. It means that the double-humped barrier may look different for different single-particle configurations. In particular, the location and width of the fission resonances will vary with the value of $\Omega$. There will be a specific pattern of fragment angular distributions associated with each band of a given $\Omega$ value. In principle one just has to find the fission resonance peaks, measure the angular distribution across the peak, and deduce $\Omega,\Gamma$.

Then one has to decide whether the resonance corresponds to the zero-order $\beta$-vibrational motion of the core or to a higher order phonon coupled to the single particle. If it is the zero-order phonon, the resonance represents the single particle state in the second well with its associated rotational band.

In practice this is not so simple, but let us see how far the exploration of single-neutron states in the second well has advanced. It will be wise to seek guidance from the cartographic works of the theorists. Figure 12 is one such map, due to Nilsson et al. [14]. The position of the second minimum is calculated from this map and found to be $\varepsilon = 0.60$. The intersection of a vertical line at $\varepsilon = 0.60$ gives the ordering of single-particle eigenvalues, shown in figure 13. The Fermi level for a shape isomer with given neutron number is determined.
isomer-ratio analysis of the results they conclude that the four spin pairs shown in Table I are the most probable candidates for the two isomers, with the two additional ones being possible also, but less probable. The main assumption required is that the spontaneous-fission cross-section ratio is representative of the total isomer cross sections. If one isomer had a decay branch back to the first well, and the other did not, the conclusions would be wrong, but this seems unlikely. Taking guidance from the Nilsson diagram, figures 12 and 13, the authors of ref. [18] reject four of the six options, being left with the $^{505}_{11/2^-}$ as the

by counting up from the bottom. It is denoted by F (for Fermi). For the two cases $^{237}_{\text{Pu}}$ and $^{233}_{\text{Th}}$ both with $N = 143$, it is the Nilsson level $[N_n \Lambda] \Omega^\Pi = [862]3/2^+$. Similarly the Fermi level for $^{231}_{\text{Th}}$ is the $[512]3/2^-$ configuration. Note the gap at $N = 144$ and the high-spin $[505]11/2^-$ state, F-3, which is responsible for a number of isomeric states in the first well at the beginning of the region of deformed rare earth nuclei [17].

In $^{237}_{\text{Pu}}$ there are two fission isomers. One with a 1.1 μs half-life, the other decaying with a 0.08 μs period. Russo et al. [18] have studied the relative population of these two isomers in compound nucleus reactions with different amounts of angular momentum brought into the compound system. They find that increased angular momentum favors the population of the 1.1 μs isomer. Using the traditional, statistical,

FIG.12 - Nilsson diagram for neutrons at large deformations [14]. Several other versions have been suggested, depending on the choice of the hexadecapole distortion parameter $\epsilon_4$, and on the radial dependence of the potential: Harmonic oscillator, Woods-Saxon [15] or a folded Yukawa potential [16]. There are significant differences between the various versions. The above example is not unique.

FIG.13 - The order of filling of neutrons in the potential, figure 12, for $\epsilon = 0.60$. Asymptotic quantum numbers are given on the left hand side. The Fermi level, F, corresponding to two nuclei that have been experimentally studied, is indicated on the right hand side ($N = 143$).
TABLE I
Possible Spin Pairs for Double Shape Isomers of $^{237}\text{Pu}$

<table>
<thead>
<tr>
<th>From measured $\lambda$-dependence of reaction yields</th>
<th>$(9/2,11/2)$ $(7/2,11/2)$ $(7/2,9/2)$ $(5/2,11/2)$ ( \rightarrow ) $(5/2,9/2)$ $(3/2,11/2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>From result above plus Nilsson diagram</td>
<td>$5/2^+,11/2^-$ ( \rightarrow ) $(3/2^-,11/2^-)$</td>
</tr>
<tr>
<td>Interpretation</td>
<td>$1.1 \text{ ms: [505]}11/2^-$ ( \rightarrow ) $0.08 \text{ ms: [862]}5/2^+$ ( \rightarrow ) $[512]3/2^-$</td>
</tr>
</tbody>
</table>

Candidate for the long lived isomer and the $[862]5/2^+$ as the most likely assignment of the short lived state. This is indeed the Fermi level in $^{237}\text{Pu}$ and the $11/2^-$ state is the F-3 level. Altogether a very consistent picture; though of course, spectroscopists working with states in the first well may have to recall the very early days of their trade to find such results satisfying.

The barriers surrounding the isomers of $^{237}\text{Pu}$ are 2-3 MeV high. That is why the life-
times are microseconds. The widths that one should find in a resonance experiment designed to reveal these states are correspondingly of the order of $10^{-9}$ eV, i.e. impossible to measure. The situation is different with the thorium isotopes. Here, the second well apparently is very shallow so that the widths become measurable, typically 10 keV or more. Figure 14 shows the result of Lynn, James and Earwaker [19] for fission induced in $^{230}\text{Th}$ by 600-1400 keV neutrons with an energy resolution of 5 keV. The fragment angular distributions in the region of the peak shows uniquely that the $\Omega$-value is one half. The various rotational states of this $\Omega = \frac{1}{2}$ band are hidden in the peak. By extremely careful high-resolution measurements [19,20] of the fragment angular distributions at the flanks and across the top of the peak, the outline of a strongly decoupled rotational band emerges.

Nature has been unkind in this case. The angular distributions are not as rich in structure as one might justly have expected with a better behaved value of the decoupling parameter (a). Figure 15 summarizes the most likely interpretation of the results for $^{231}\text{Th}$. In addition to the $\Omega = \frac{1}{2}^-$ band which fits very naturally as the F-1, $[510]1/2^-$ configuration at neutron number, $N = 141$, there is evidence of a $\Omega^u = 3/2^-$ resonance, i.e. the Fermi level itself $[512]3/2^-$ (see figures 12 and 13).

The uniqueness and in particular the
purity of the asymptotic quantum numbers, assigned in this way, remains open to question. In figure 12 one sees how states with the same value of $\Omega^\pi$ repel each other. This is equivalent to mixed asymptotic quantum numbers. In the vicinity of $E = 0.6$, the $[510]1/2^-$ and the $[750]1/2^-$ states may be mixed, for example. As a result the decoupling parameter is not well determined theoretically.

The neutron induced fission of $^{232}\text{Th}$ is another case where the spectroscopy of fission resonances has been pushed quite far by Andro- senko et al. [21]. Here, the situation is in a way the inverse of the $^{231}\text{Th}$ situation. There are no beautiful peaks in the fission excitation function like in figure 14, but the fragment angular distributions show very pronounced structure, figure 16. Note for example the dramatic change occurring between 1.30 MeV and 1.35 MeV neutron energy, i.e. in an interval of only 50 keV. The Moscow-Obninsk group has boldly applied a resonance analysis to the results and proposed two not very different versions to explain them: Five to six rotational bands in the second well with specific values of $\Omega^\pi$ are responsible for the total fission cross section and the angular distributions. Figure 17 shows...
Possible Neutron States in Second Well from Fission Resonances and Fragment Angular Distributions in \( ^{232}\text{Th} + n \)

Androsenko et al.

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>Assignment</th>
<th>Energy (MeV)</th>
<th>Assignment</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.8</td>
<td>5/2+</td>
<td>5.9</td>
<td>1/2+</td>
</tr>
<tr>
<td>6.0</td>
<td>5/2-</td>
<td>6.0</td>
<td>3/2+</td>
</tr>
<tr>
<td>6.15</td>
<td>3/2-</td>
<td></td>
<td></td>
</tr>
<tr>
<td>F</td>
<td>F+2</td>
<td>?</td>
<td>?</td>
</tr>
</tbody>
</table>

In conclusion, let us mention a few specific lines of attack where one can hope for progress in a not too distant future.

a. Doubly Even Nuclei.

1. A determination of the quadrupole moment of the shape isomer would be a spectacular contribution.

2. Rotational bands in isomers in addition to \(^{240}\text{Pu}\) could be studied.

3. There are several more candidates for K-isomerism, notably \(^{236}\text{Pu}, \(^{242}\text{Cm}\) and \(^{244}\text{Cm}\), where a determination of the energy gap could be attempted. The problem is that of finding the \(0^+\), ground state fission isomer, which is likely to decay within picoseconds. Another interesting problem is to find out whether the K-isomers decay by gamma emission to ground in the second well or directly by spontaneous fission.

4. More precise determinations of the K,\( \kappa \) quantum numbers of the vibrational resonances would be useful.

b. Odd-A Nuclei.

5. The energy difference between the two isomers in \(^{237}\text{Pu}\) would be interesting to know. It could lie anywhere between zero and several hundred keV.

6. There are presumably more double isomers

their proposed decomposition of the measured fission probability curve. Each band is located at the lowest peak (or shoulder). In Table II neutron energy is translated to excitation energy above the \(^{233}\text{Th}\) ground state (first well), and the \( \Omega^\pi \) values proposed in the two versions are listed. The lowest band is quite near in energy to the \( 1/2^- \) band in the neighbouring \(^{231}\text{Th}\), figure 15. One may now consult figures 12 or 13 to see if the experimental \( \Omega^\pi \) values correspond to those predicted to lie near the Fermi surface of \(^{233}\text{Th}\). Three out of five cases work well, particularly in case b; the remaining two do not. It is easy to suggest these states to be due to octupole or quadrupole vibrations coupled to the pure single-particle states. The phonon energies are a bit low though, 0.2 - 0.25 MeV. In any case, this approach will be sure to explain everything and predict nothing. One could also take the works of other cartographers and see if their predictions are more amicable, see caption to figure 12. All in all, the study [21] of \(^{233}\text{Th}\) represents a promising beginning and one can hope for further progress with this nucleus.

IV - CONCLUDING REMARKS

The spectroscopy of shape isomers is still in its infancy. Although the baby develops promisingly, it is premature to make status with respect to those features of special symmetry which were discussed in the introduction. They are expected to reveal themselves in the level structure, for example through a relatively weak coupling between different degrees-of-freedom.
SHAPE ISOMERS

Besides the $^{237}\text{Pu}$ case in the odd-N isotopes. Similarly, the identification of double isomers in odd-Z nuclides would open the discussion of the theoretical proton level diagram.

7. The band structure associated with the $^{233}\alpha$ resonances could perhaps be studied more closely in order to confirm the assignments, derive moments of inertia, etc.

8. Application of perturbed fragment angular-correlation techniques may open the field of magnetic moment measurements and perhaps also of quadrupole moments.

c. Doubly Odd Nuclei.

9. Vibrational type resonances have been found in $^{232}\text{Pa}$. [22]. So far, it has not been possible to assign quantum numbers.

10. Double fission isomers may also be found in doubly odd nuclei.

d. New Regions of Shape Isomers

11. A significant next step would be the discovery of a new island of shape isomers. The region around $N = 118$ and $Z = 84$ is the most promising candidate. Here, the shape isomers are expected to decay by $\gamma$-emission [23]. This is a difficulty, but the cross sections could be higher than in the transuranium region, because compound nucleus fission is less likely to exhaust the reaction cross section.

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H. NIFENECKER (Saclay)

How can you explain that the life times of the excited states in the second well are longer than the ground state life time?

S. BJØRHOLM (Niels Bohr Inst.)

The excited state has a higher energy because energy is required to break a pair and form a two-quasiparticle configuration with definite quantum numbers $K^\pi$. This is true for all shapes, so the effective barrier seen by the excited state is as high as the one seen by the ground state. Besides, the inertia associated with the barrier penetration is likely to be higher for an unpaired system than for a completely paired system. This would make the life time longer for the two-quasiparticle state.

J. JAŚTRZEBSKI (Pologne)

Can you comment about the non existence of Np spontaneously fissioning isomers?

S. BJØRHOLM (Niels Bohr Inst.)

I think the shape isomers are there, but they decay by penetration of the inner barrier and emission of gamma rays.

E.R. HILF (Darmstadt)

The theoretical prediction (see CROOKE et al. Nucl. Phys. A129 (1969) 513) of the small island of shape isomers near Ra resulted from the interplay between the surface and curvature tension $\kappa$ in the liquid drop model which yields a flat at the top or even double humped barrier for nuclei near Ra, the lighter (heavier) ones having only one saddle of constricted (ellipsoidal) shape. Then R.W. HASSE (Ann. Phys. 68 (1971) 377) could show that this effect of the LDM is not spoiled by the additional terms of the droplet model. In that area at least the D.M. flattens the top of the barrier and thus allows for the shell effects digging a second well despite the steep barrier, if only $\kappa$ is sufficiently large, at least the 9 MeV of V. GROOTE barrier fit. However B. MYERS and W. J. SWIATECKI (priv. Comm. at this Conf.) now end up with $\kappa \geq 0$ MeV. Thus this island of shape isomers may not exist.