γ

UCRL- 97800
PREPRINT

Conversion-electron experiment to characterize the decay of the ^{237}Np shape isomer

E. A. Henry
J. A. Becker
R. W. Bauer
D. G. Gardner
D. J. Decman
R. A. Meyer
N. Roy
K. E. Sale

This paper was prepared for presentation at
O/E/LASE '88
Los Angeles, CA
January 10-15, 1988

December 8, 1987

Lawrence Livermore National Laboratory

Conversion-electron experiment to characterize the decay of the ^{237}Np shape isomer

E.A. Henry, J.A. Becker, R.W. Bauer, D.G. Gardner,
D.J. Decman, R.A. Meyer, N. Roy, and K.E. Sale

Lawrence Livermore National Laboratory
Livermore, CA 94550

UCRL--97800

DE88 004402

ABSTRACT

Conversion electrons from the decay of low-lying levels of ^{237}Np have been measured to detect the population of these levels by gamma-ray decay of the ^{237}Np shape isomer. Analysis of the 208-keV transition L conversion-electron peak gives an upper limit of about 17 μb for the population of the $3/2^-$ 267-keV level in ^{237}Np from the shape isomer decay. Model calculations are compared with the measured limit. Improvements are suggested for this experiment.

1. INTRODUCTION

Nuclear isomerism plays a key role in current conceptual designs of a gamma-ray laser.[1] In these designs, the isomeric state stores a large amount of energy that would be released to provide a coherent beam of electromagnetic radiation. Nuclear isomerism results from one or a combination of nuclear structure factors: a small energy difference between nuclear energy levels, a large spin change between levels, K-forbiddenness of the transition connecting levels (in the case of deformed nuclei), or a difference in nuclear shape associated with the levels of interest. Of these factors, nuclear shape isomerism is the least studied, with virtually no systematic experimental information existing on isomer occurrence as a function of nuclear mass and charge, the isomer decay modes, isomer spin and parity, or other properties.

Shape isomers are the isomers that result when the nuclear potential exhibits a second energy minimum at a deformation, or shape, different from that of the normal nuclear ground state. Low-spin shape isomers have been identified only in the actinide nuclei where they are known as fission isomers because they were discovered and studied mainly through their fission decay mode.[2] In the light actinide nuclei, however, it is believed that gamma-ray decay is the dominant de-excitation mode of the shape isomers. We are studying the gamma-ray decay of the shape isomers in the light actinide nuclei to extend the systematic knowledge of their properties, and to develop techniques and strategies to identify shape isomers in nuclei where the fission decay mode does not occur.

The nucleus ^{237}Np is known from fission studies to have a shape isomer with a half-life of 45 ns.[3] Based on the fission half-life systematics of Metag,[4] this isomer decays mainly by gamma-ray emission, with the gamma-ray branch being about 99.9%. The isomeric level, the ground state in the deformed second minimum, is at an excitation of 2.85 ± 0.4 MeV. We have previously searched for the high energy primary gamma rays from this isomer using the ^{238}U(p,2n)^{237}Np reaction at E_p = 12 MeV. With this reaction the isomer is formed with a total cross section calculated to be 40 μb. We obtained an upper limit of 5.3 μb (2 sigma) for any single gamma-ray with an energy between 1600 and 3300 keV associated with the decay of the isomeric state back to the first well.[5] Subsequent calculations suggested that the decay of the shape isomer by a single intense gamma ray might not be the dominant de-excitation mode. Rather, up to ten or more primary transitions in that energy range might depopulate the isomeric level, with virtually none of them having a cross section of as much as 5.3 μb. These calculations suggested however, that the ^{237}Np shape isomer would decay up to half the time to certain low-lying levels in the first well, depending on the isomer spin and parity. These low-lying levels generally have short half-lives or decay promptly. Thus a 45 ns component in their decay curve would indicate that they are probably populated by the gamma-ray decay of the shape isomer. The experiment reported here was designed to search for this process.

2. EXPERIMENTAL PROCEDURE

Conversion-electron spectroscopy is the best method[6] to study low-energy transitions when fission is the main competing reaction, as it is in this experiment. Because fission fragments have a high kinetic energy, they recoil out of the focal volume of a focusing device like a solenoidal electron spectrometer and their radioactivity does not contribute significantly to the background. This experiment exploited the capabilities of the Lawrence Livermore National Laboratory's solenoidal conversion-electron spectrometer, which is described in detail in ref. 6.

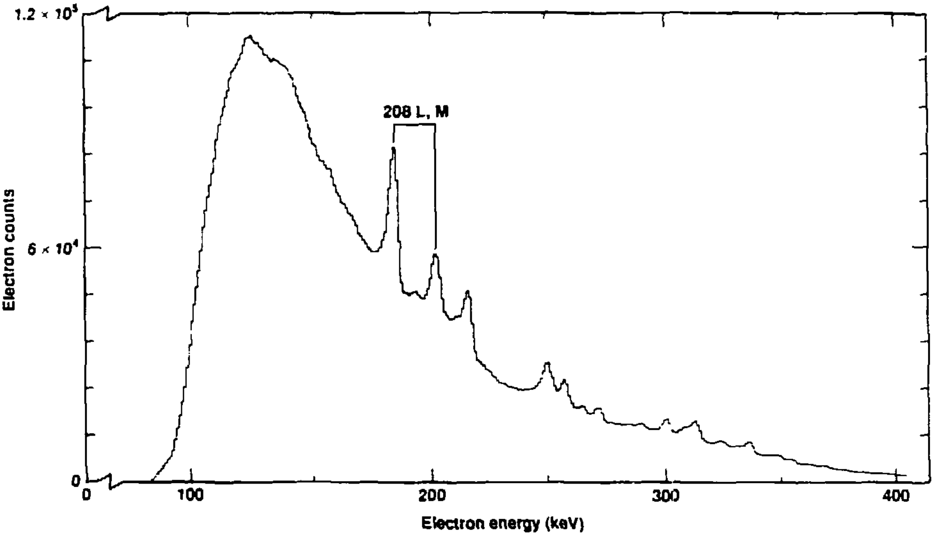

Figure 1. Time-integrated electron energy spectrum. The 208-keV transition L and M conversion electron peaks originating from the 267-keV level in ^{237}Np are identified. *The peaks observed in this spectrum arise mainly from the* direct reactions.

A thin target of uranium oxide (300 $\mu g/cm^2$ of ^{238}U) mounted on a carbon foil (30 $\mu g/cm^2$) was bombarded with 12 MeV protons from the Los Alamos FN tandem Van de Graaff accelerator. The target was oriented so that the beam impinged nearly parallel to the target foil to increase the reaction rate and, in our geometry, facilitate detection of electrons and fission fragments. The proton beam was bunched with a pulse width of about 2 ns and a repetition rate of 156 kHz. Event time information was obtained relative to the beam pulse on target.

Conversion electrons were transported by the solenoidal magnetic field through a baffle system to a 200 mm^2 Si(Li) detector, the energy dispersive element in the spectrometer. The magnet current was swept so that electrons with energies in a selected energy range could be detected with reasonable efficiency. The absolute efficiency for electron detection was obtained by counting a calibrated source of ^{152}Eu. The dead time of the electron detector electronic system was determined by using a random pulser signal as an auxiliary input into the preamplifier. A 100 mm^2 Si(Li) fission fragment detector monitored the prompt fission fragments and, together with the measured fission cross section for this reaction, determined the number of prompt fissions that occurred in the target. This value served to calibrate the other reaction cross sections.

Low-energy transitions of interest in ^{237}Np include the 208-keV transition between the $3/2^-$ level at 267 keV and the $5/2^-$ level at 59 keV, and the 96-keV transition between the $11/2^+$ level at 129 keV and the $7/2^+$ level at 33 keV. Initially, the K conversion electrons from the 208-keV transition were detected at 89 keV, but the L conversion electrons from the 96-keV transition could not be observed at 74 keV. However, the peak-to-background ratio was small throughout this energy region because of the intense delta electron (atomic electrons ejected by the projectile) contribution to the background at low energies. Therefore, the spectrometer transmission was adjusted to detect the 208-keV transition L conversion electrons since the delta electron background falls rapidly with increasing energy. The electron spectrum was digitized and accumulated as a two dimensional array in a large on-line memory as a function of time after the beam pulse, and also stored on magnetic tape for later analysis. Data were written to tape approximately every 16 h so that corrections could be made for small energy drifts in the detector system. Data were accumulated for 82 h and 1.41 x 10^{10} fissions occurred in the target during that time.

3. DATA ANALYSIS

The 208-keV transition L and M conversion-electron peaks are clearly visible in the time integrated spectrum shown in Fig. 1. This transition, and others visible in the spectrum, result mainly from direct population by the (p,2n) reaction. If a portion of the counts from these transitions is due to decay of the ^{237}Np shape isomer, that portion will be

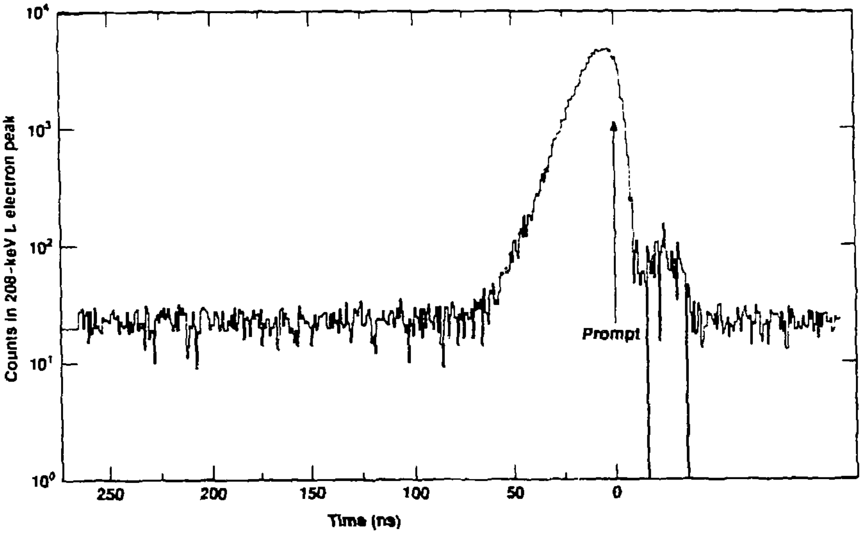

Figure 2. Decay with time of the 208-keV transition L peak. A time spectrum from a nearby background region has been subtracted and twenty counts added to each channel. The 267-keV level from which this transition originates has a 5.2 ns half-life that is clearly observed. No 45 ns component, indicative of population from the isomer decay, is observed in this spectrum.

characterized by a 45 ns half-life, the half-life of the isomer. Shown in Fig. 2 is the time distribution of the 208-keV transition L peak after 10 h of data accumulation. A background has been subtracted and twenty counts added to all channels to display the statistics at late times. The 208-keV transition originates from a level[8] that has a half-life of 5.2 ± 0.2 ns. A preliminary value for that half-life extracted from the data of Fig. 2 is 5.4 ns, in excellent agreement with the accepted value. A component with a 45 ns half-life cannot be seen in this spectrum or in the summed time spectrum.

In order to examine the time distribution of the 208-keV transition L peak in detail, the electron spectrum was projected from the two-dimensional data for two time periods: 67 to 157 ns and 157 to 255 ns after the beam pulse. After 67 ns, all but 0.019% of the 267-keV level has decayed. Thus only about 200 counts occur in the 208-keV transition L peak during the 67-157 ns time period unless the shape isomer decays to the 267-keV level. The electron spectrum for the first time period is shown in Fig. 3. As can be seen, several weak long-lived peaks in the region of interest complicate the data interpretation. Careful fitting of the spectrum indicates that the multiplet near 186 keV consists of two unidentified peaks, with one 1.4 keV below and the other 1.4 keV above the energy of the 208-keV transition L peak. These two peaks are also observed in the later time period. The ratio of counts in the two time periods indicates that the half-lives of these two peaks is greater that 60 ns. No peaks are detected in this energy region when 6.4 μs has elapsed after the beam pulse. From these considerations it is concluded that a 45 ns component has not been observed for the 208-keV transition L peak.

An upper limit for the decay of the shape isomer to the 267-keV level can be calculated from an analysis of the background counts in the vicinity of the 208-keV transition L peak. This upper limit is obtained by combining the square root of the background summed over 1.75 keV, the fraction of the peak occurring in this energy range, the fraction of decays occurring during the 67-157 ns time period, the fraction of the transitions occurring via L conversion, the electronic system live time, the branching ratio of the 208-keV transition from the 267-keV level, the electron spectrometer efficiency, the fission fragment detector efficiency, and the known prompt fission cross section. The two sigma upper limit obtained is 17 μb. This value includes the combined two sigma uncertainty of 24% in the central values of the parameters described above.

It must be emphasized that the upper limit deduced above is valid only for a truly structureless background. The fact that long-lived small peaks exist in the spectrum during the important time period makes it difficult to rule out the possibility of some isomer feeding to the 267-keV level at about the value deduced

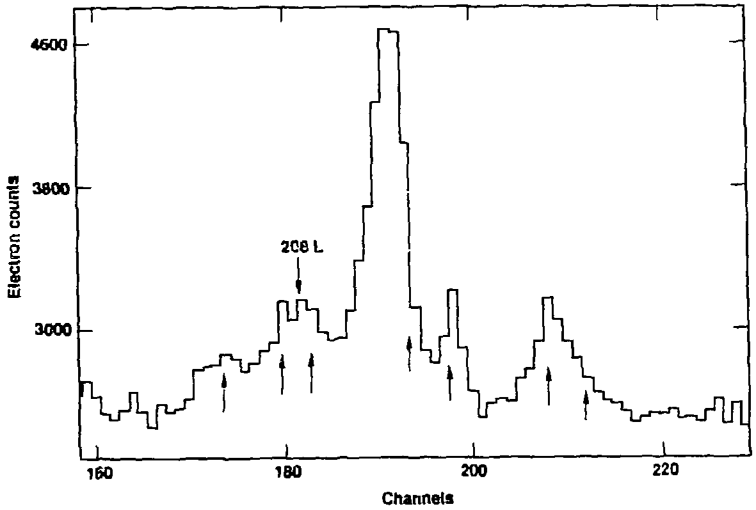

Figure 3. Electron energy spectrum for a time period 67 to 156 ns after the beam pulse. Delayed peaks of unknown origin are indicated by arrows below the histogram. Two such peaks bracket the expected position of the 208-keV transition L electron peak. Both peaks have a half-life of greater than 60 ns. These small peaks make identification of low-level population of the 267-keV level difficult.

above for the limit. Further analysis is required to elucidate the limitations of these data.

4. CALCULATIONS

Calculations have been performed using the STAPLUS code[9] to study the gamma-ray decay of the shape isomer. It is assumed that the population of the isomeric state "tunnels" through the potential barrier by mixing with states at nearly the same energy in the normal ground state potential well. The admixed isomeric level can then decay by electromagnetic transitions in the normal way through low-lying discrete levels to the ground state. The STAPLUS code allows the population of one or more levels within an arbitrary energy interval and with an arbitrary spin and parity. The gamma-ray decay is then followed in two energy regions. Near the ground state the code accepts discrete energy levels with branching ratios to lower-lying levels as input information. Above the discrete levels up to the arbitrarily populated level, the code uses a statistical description of the nuclear level density. The primary gamma ray decays out of the populated level, and gamma rays between the statistical levels, are assumed to be mainly dipole in nature. The transition strength of these dipole transitions is adjusted to match experimental neutron-capture gamma-ray data.

The ^{237}Np shape isomer decay calculations were carried out with the populated level at three different energies (2.4, 2.8, and 3.2 MeV) since the isomer energy is uncertain by 0.4 MeV. Similarly, since the isomer spin and parity are not known, the populated level was assigned a spin with values from 1/2 to 15/2, with the parity being either positive or negative, in successive calculations. Two sets of discrete levels, each containing 110 energy levels, were used in the calculations. These two sets differed only in the completeness of the branching ratio information. The first set included branching ratios reflecting the assumption that levels in a rotational band decayed to lower levels of the same band until the bandhead level was reached. The decay of the bandhead level to levels in other rotational bands was then calculated using a rotational model. The second set of discrete levels included all the experimentally known branching ratios. The branching ratios of about 20% of the levels were modified in the second set compared to the first.

As mentioned briefly in the introduction, a calculation of the primary gamma rays, those emitted from the original populated level, showed that virtually none had a production cross section of 5.3 μb or greater. The only situation where the calculated cross section for a primary gamma ray was larger than 5.3 μb occurred with the isomer spin assumed to be 1/2 and the isomer energy of 2.4 MeV. In that instance the primary gamma rays to the two lowest 3/2 levels had cross sections of about 5.8 μb.

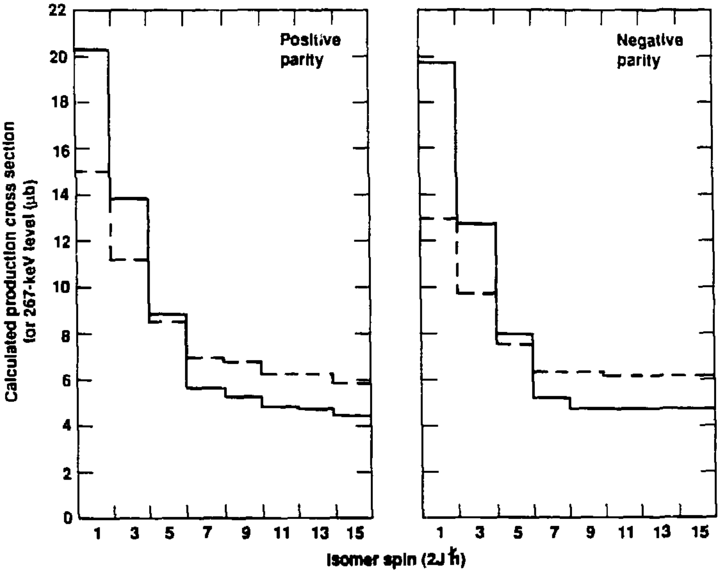

Figure 4. Calculation of the production cross section of the 267-keV level from the shape isomer decay as a function of the isomer spin and parity. The calculation of the cross section is done with two sets of discrete level information: the set with calculated branching ratios (dashed histogram), and the set that contains the known experimental data (solid histogram). In these calculations the production cross section for the shape isomer is assumed to be 40 μb.

A number of primary gamma rays had cross sections of greater than 1 μb for all energy, spin, and parity combinations. From the comparison of the calculated gamma ray cross sections to the measured upper limit it is concluded that the decay of the shape isomer is fragmented over a number of primary gamma rays, and that fluctuations from the average cross sections calculated here are insufficient to cause any one transition to have a production cross section of greater than 5.3 μb.

While the calculation of primary gamma-ray cross sections depends only on the statistical level density and the discrete levels, the calculation of the population of a low energy discrete level depends significantly on the branching ratio information. Fig. 4 shows the population of the 267-keV level (which is depopulated mainly by the 208-keV transition) when the two different sets of discrete level information are used. The cross section for population of the 267-keV level by isomer decay is about 50% of the isomer population (assumed to be 40 μb) when the isomer spin is 1/2 and the discrete level set containing the measured data is used. The cross section of Fig. 4 is to be compared with the experimental upper limit of 17 μb (assuming other delayed transitions do not mask evidence of real population by the isomer).

5. CONCLUSIONS

The present experiment is the first reported that attempts to identify shape isomer gamma-ray decay by characterizing its population of low-lying discrete levels. Analysis of data so far indicates that the cross section upper limit for population of the 267-keV level is near the cross section that a calculation would predict. Other electron peaks observed in the experiment have not yet been subjected to detailed analysis and may yield information about the shape isomer decay.

A number of improvements could be made in the present experiment to improve its sensitivity substantially:

1. Replace the oxide target with a metallic target. Previous experiments with a metallic uranium target mounted on a carbon foil indicates that there is less delta electron background than experienced in this work. A qualitative observation during the present experiment supports this indication. The delta electron rate dropped continually during the experiment. At the conclusion of the experiment it was observed that the target, originally a yellow uranium oxide, had been largely reduced to the black metallic state at the spot where the proton beam impinged on it. With a greatly lowered delta electron background, a future

experiment could emphasize the 208-keV transition K conversion-electron peak. This could improve the sensitivity by up to a factor of five.

2. Reduce the general long-lived background and measure electron spectra for a longer time after each beam pulse. One cause of long-lived background is the thermalization and capture of neutrons from the target and beam stop. Shielding could be optimized to reduce this background and the beam pulse rate could be decreased while maintaining the same average beam current. Data from the present experiment indicates that during the first 6.4 μs after a beam pulse, the general background decreases about 40%. Decreasing the pulse rate by a factor of four might reduce the general background by as much as a factor of 2.5 if that trend continues. Longer counting intervals after each beam pulse would also allow better characterization of the long-lived background.

3. Decrease the electron energy range swept by the spectrometer field to study a particular transition. The average counting efficiency over a narrower electron energy range could be easily increased by a factor of two or more.

4. Increase the beam current. Step 2 above will increase the electronic system dead time. However, in the present experiment the dead time was only 4.4%. Previously experiments of this type have been carried out with dead times as high as 30% or more. The present experimental setup could therefore easily accommodate an increase in beam current by up to a factor of two.

Combining these factors, it seems possible that the sensitivity of this type of experiment could be increased by a factor of at least three over the present work.

This work was performed by the Lawrence Livermore National Laboratory supported in part by the United States Department of Energy under contract #W-7405-ENG-48, and in part by IST/SDIO, directed by the Naval Research Laboratory.

6. REFERENCES

1. G. C. Baldwin, J. C. Solem, and V. I. Gol'danskii, "Approaches to the development of gamma-ray lasers," Rev. Mod. Phys. 53, 687 (1981).

2. S. Bjornholm and J. E. Lynn, "The double-humped fission barrier," Rev. Mod. Phys. 52, 725 (1980).

3. E. Migeco, G. Russo, R. DeLeo, and A. Pantaleo, "Fission isomer of $^{237}Np^{m}$," Phys. Rev. C 16, 1919 (1977).

4. V. Metag, "Systematics of fission isomer half-lives", Nucleonica 20, 789 (1975).

5. E. A. Henry, J. Kantele, D. J. Decman, J. A. Becker, W Stoeffl, and L. G. Mann, "Search for the gamma-ray decay of the ^{237}Np fission isomer," Nuclear Chemistry Division Annual Report FY 86, UCAR 10062/86, p2-48 (1986).

6. W. Stoeffl and E. A. Henry, "An electron spectrometer with a superconducting magnetic transporter for in-beam use," Nucl. Instr. Meth. 227, 77 (1984).

7. J. R. Boyce, T. D. Hayward, R. Bass, H. W. Newson, E. G. Bilpuch, and F. O. Purser, "Absolute cross sections for proton-induced fission of the uranium isotopes," Phys. Rev. C 10, 231 (1974).

8. Y. A. Ellis-Akovali, "Nuclear data sheets for A = 237," Nucl. Data Sheets 49, 181 (1986).

9. STAPLUS is a version, under development, of the STAPRE statistical model nuclear reaction code by M. Uhl and B. Strohmaier described in: H. Vanoch, "Users manual for the code STAPRE as implemented at Lawrence Livermore National Laboratory," UCID-19549, Lawrence Livermore National Laboratory (1982).