NEW RESULTS ON FISSION CROSS SECTIONS IN ACTINIDE NUCLEI USING THE SURROGATE RATIO METHOD AND ON CONVERSION COEFFICIENTS IN TRIAXIAL STRONGLY DEFORMED BANDS IN ^{167}Lu FROM ICE BALL AND GAMMASPHERE*

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The surrogate ratio technique is described. New results for neutron induced fission cross sections on actinide nuclei, obtained using this technique are presented. The results benchmark the surrogate ratio technique and indicate that the method is accurate to within 5% over a wide energy range. New results for internal conversion coefficients in triaxial strongly deformed bands in ^{167}Lu are also presented.

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1. Introduction

Direct measurements of neutron-induced reactions on unstable nuclei pose significant experimental challenges. For example, the half-life of ^{237}U is one week, thus making a direct measurement extremely difficult. In many

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cases the surrogate technique can be utilized to overcome the problems inherent in direct measurements of compound-nuclear reaction cross sections. In the surrogate method, the compound nucleus relevant to the reaction of interest ("desired reaction") is produced via a direct reaction, but using stable beams and targets. A measurement of the decay probabilities of this compound nucleus can then be combined with a calculation of the formation of the compound nucleus in the desired, neutron-induced, reaction to yield the sought-after cross section. In the surrogate ratio method, a new variant of the standard surrogate approach, measurements are carried out in order to determine the ratio of two cross sections. Knowledge of one of these cross sections then allows one to infer the other. The ratio technique eliminates many of the problems encountered in the absolute surrogate technique; mainly these problems are due to target contamination issues.

The first measurement using the surrogate ratio technique utilized the Silicon Telescope Array for Reaction Studies (STARS) detector array, coupled to the Yale Rochester Array for SpecTroscopy (YRAST Ball) at the Wright Nuclear Structure Laboratory at Yale, to extract the cross section ratio $\frac{\sigma[^{236}\mathrm{U}(n,f)]}{\sigma[^{238}\mathrm{U}(n,f)]}$ and $\frac{\sigma[^{235}\mathrm{U}(n,f)]}{\sigma[^{237}\mathrm{U}(n,f)]}$ via the (d,pf) and $(d,d'f)$ reactions, respectively, on ^{236}U and ^{238}U targets. More recent results on the ratio $\frac{\sigma[^{235}\mathrm{U}(n,f)]}{\sigma[^{233}U(n,f)]}$, obtained via $(\alpha,\alpha' f)$ reactions on ^{236}U and ^{234}U targets, carried out using STARS at Lawrence Berkeley National Laboratory are also presented. These results serve to benchmark the technique and show that the surrogate ratio method can be accurate to $\sim 5\%$ or better over an excitation energy range of 7 to ~ 25 MeV. In addition, in the second half of this paper very recent results on conversion coefficient measurements of the triaxial strongly deformed bands in ^{167}Lu are presented.

2. New results on fission cross sections from the surrogate ratio method

Neutron-induced cross sections play important roles in a wide variety of nuclear and astrophysical phenomena including for example, fission and the production of heavy elements in stellar processes. However, for short-lived isotopes it is often difficult or impossible to measure such cross sections directly. For example, the target may be impossible to construct, or may be highly radioactive leading to a very intense background. The surrogate technique, first pioneered in the 1970's [1–3], is an attempt to deduce such cross sections by combining a calculation with an alternative or "surrogate" reaction that provides information on the decay of the compound-nucleus through which the desired reaction proceeds.

Central to the surrogate technique is the notion that the decay of the compound nucleus is independent of the manner by which it was produced. However, the decay probabilities that enter into a Hauser–Feshbach description of a compound-nuclear reaction are known to depend on the excitation energy and on the spin and parity of the states populated in the compound nucleus (see the discussion in Ref. [4]). Since the fusion of a neutron with a target nucleus is likely to result in a compound-nuclear spin-parity population that is different from the one produced in the surrogate reaction, the effects of this "spin mismatch" should be accounted for in a complete treatment of a surrogate experiment. Almost all applications so far, including the ones presented here, ignore this issue. This raises several questions: can a surrogate reaction produce a compound nucleus with a spin-parity distribution that is similar to the one produced in the neutron-induced reaction? What effect does a mismatch in the angular momentum distributions of the direct and surrogate reactions have on the inferred decay probabilities? How large a mismatch can one tolerate and still obtain reasonable results with the surrogate method? Furthermore, the question of how the highly-excited nuclear system that is created in the surrogate reaction equilibrates (*i.e.* becomes a compound nucleus) has not been investigated yet. The likelihood of particle emission from the highly-excited nucleus prior to equilibration, which would invalidate the surrogate analysis, needs to be estimated.

To address these and other questions two things are needed (a) improved theoretical modeling of the reaction mechanisms and (b) high-quality surrogate reaction data on a variety of systems where the direct measurements have been made, to benchmark the technique. Significant progress on both theoretical [4,5] and experimental [6–11] fronts has been made over the last several years. Here the newly developed surrogate ratio [4,7,10–12] technique will be briefly described and new data on the $^{234}\mathrm{U}(\alpha,\alpha' f)/\ ^{236}\mathrm{U}(\alpha,\alpha' f)$ ratio, surrogate for the well-known $^{233}\mathrm{U}(n,f)/\ ^{235}\mathrm{U}(n,f)$ reaction will be presented.

As a specific example, consider a measurement of the neutron-induced fission cross section $\sigma[^{237}\mathrm{U}(n,f)](E)$, over a wide range of neutron energies E, from a few hundred keV up to many MeV. Since the half-life of $^{237}\mathrm{U}$ is only about one week the construction of a target is very difficult. A possible surrogate reaction is $^{238}\mathrm{U}(\alpha,\alpha' f)$. The cross section of interest, $\sigma[^{237}\mathrm{U}(n,f)](E)$, can be written as the product of two terms:

$$\sigma\left[{}^{237}\mathrm{U}(n,f)\right](E) = \sigma_{(\mathrm{abs})} P(f)[E]\,.$$

Here the absorption cross section, $\sigma_{(\mathrm{abs})}$, the formation cross section of $^{238}\mathrm{U}$ from $n+^{237}\mathrm{U}$, is obtained from an optical model calculation. The fission probability $P(f)$, is the experimentally measured quantity in the surrogate reaction and is given by

$$P(f) = \frac{N[\,^{238}\mathrm{U}(\alpha, \alpha' f)]}{N[\,^{238}\mathrm{U}(\alpha, \alpha')]}\,.$$

where $N[\,^{238}\mathrm{U}(\alpha, \alpha' f)]$ is the number of fission events measured in coincidence with scattered alpha particles and $N[\,^{238}\mathrm{U}(\alpha, \alpha')]$ is the total number of scattered alpha particles. The measured energy of the scattered alpha particle is used to deduce the excitation energy in the composite system and hence the equivalent neutron energy.

In practice applying this technique often presents significant challenges. For example any impurity in the target material can make an experimental determination of the denominator in the above equation extremely difficult to determine. Undetected charged particle or neutron emission may also affect the measurement, leading to an error in the excitation energy of the system.

The surrogate ratio technique can overcome these difficulties by expressing the measurement as a ratio of cross sections for two similar nuclei. For example, one can experimentally measure the fission probabilities for both $^{236}\mathrm{U}(\alpha, \alpha' f)$ and $^{238}\mathrm{U}(\alpha, \alpha' f)$ which yields

$$\frac{\sigma[^{237}\mathrm{U}(n, f)]}{\sigma[\,^{235}\mathrm{U}(n, f)](E)} = \frac{\sigma_{(\mathrm{abs})} P[\,^{238}\mathrm{U}(\alpha, \alpha' f)]}{\sigma_{(\mathrm{abs})} P[^{236}\mathrm{U}(\alpha, \alpha' f)](E)}\,.$$

When evaluating this expression one must correct of course for different beam fluxes, target thickness, *etc.*, and for the (similar) absorption cross sections of the two nuclei. As can be seen the troublesome denominator $N(\alpha, \alpha')$ in P (fission) cancels in the ratio. Thus we are left with the simple expression for the ratio of cross sections:

$$\frac{\sigma[\,^{237}\mathrm{U}(n, f)]}{\sigma[^{235}\mathrm{U}(n, f)](E)} = \frac{N[^{238}\mathrm{U}(\alpha, \alpha' f)]}{N[^{236}\mathrm{U}(\alpha, \alpha' f)](E)}.$$

Further details of the surrogate ratio method can be found, for example, in references [4, 7–12].

The first test of the surrogate ratio method was carried out using the $(d, d' f)$ and (d, pf) reactions on ^{236}U and ^{238}U targets [7, 8]. In this experiment the ratio, $\frac{N[\,^{238}\mathrm{U}(d,pf)]}{N[^{236}\mathrm{U}(d,pf)]}$ was measured in order to determine the well-known cross section ratio $\frac{\sigma[\,^{238}\mathrm{U}(n,f)]}{\sigma[\,^{236}\mathrm{U}(n,f)]}$ and serves to benchmark the technique. Furthermore, the $(d, d' f)$ experiments yielded the ratio $\frac{\sigma[^{237}\mathrm{U}(n,f)]}{\sigma[^{235}\mathrm{U}(n,f)]}$, thus allowing one to extract information for the unknown $\sigma[\,^{237}\mathrm{U}(n, f)]$ cross section (at all but the lowest energies) using the known values for $\sigma[\,^{235}\mathrm{U}(n, f)]$.

For this experiment the deuteron beam, with energies of 24 and 33 MeV, was delivered by the ESTU Van de Graaff accelerator at Yale University. Scattered light ions (deuterons or protons) were detected using the Silicon Telescope Array for Reaction Studies (STARS) spectrometer which was developed by Lawrence Livermore National Laboratory. It its standard configuration, STARS consists of two CD type SiLi detectors, configured as a ΔE–E particle telescope. The thickness of the ΔE detector was $\sim 140\,\mu$m, while that of the E detector was $\sim 900\,\mu$m. To provide additional stopping power two E-detectors, one directly behind the other were utilized for the Yale experiment. Both the ΔE and E detectors were segmented into 24 rings and 8 sectors. In the Berkeley experiment an additional segmented CD detector, placed at backward angles was used to detect fission fragments in coincidence with scattered light charged particles. Additional details on the experimental setup can be found in [7, 8, 12].

The results of this first experiment are summarized in Fig. 1. Although the error bars are large, one can immediately see the promise of the ratio technique. In particular, the (d, pf) data are in good agreement with the benchmark ratio (upper dashed line in Fig. 1) for the known $\frac{\sigma[\,^{238}\mathrm{U}](n,f)}{\sigma[\,^{236}\mathrm{U}](n,f)}$ cross section ratio.

Encouraged by this result a campaign of experiments has been carried out with STARS, now located at the 88-inch cyclotron at Lawrence Berkeley National Laboratory. These experiments are designed to further benchmark

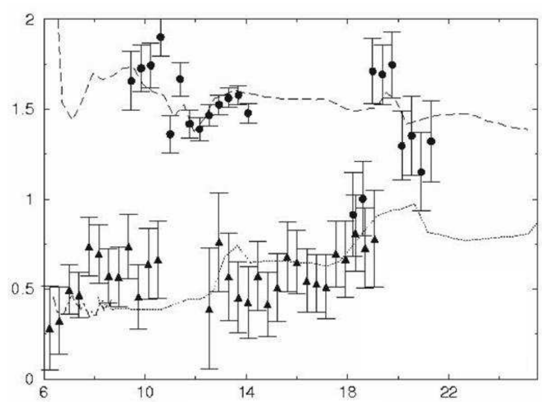

Fig. 1. The first test of the surrogate ratio method, adapted from [7]. The top set of data (circles) is the ratio of $\frac{N(^{238}\mathrm{U}(d,pf))}{N(^{236}\mathrm{U}(d,pf))}$, surrogate for $\frac{\sigma[^{238}\mathrm{U}](n,f)}{\sigma[^{236}\mathrm{U}](n,f)}$ and is compared to the accepted value for the ratio extracted from the databases (dashed line). The lower data (triangles), the ratio of $\frac{N(^{238}\mathrm{U}(d,df))}{N(^{236}\mathrm{U}(d,d'f))}$, is surrogate for the unknown $\frac{\sigma[^{238}\mathrm{U}](n,f)}{\sigma[^{236}\mathrm{U}](n,f)}$ ratio of cross sections. The x-axis shows the excitation energy in MeV.

the ratio technique, to determine its limitations (investigating, for example, the effect of the "spin mismatch" on the results [13]), and to extract new information on cross sections of interest.

The preliminary results of one of the most recent experiments are summarized in Figs. 2 and 3. Full details will be provided in [14]. The aim of the experiment was to use the $(\alpha, \alpha' f)$ surrogate reaction on ^{236}U and ^{234}U targets to measure the known cross section ratio, $\frac{\sigma[^{235}\mathrm{U}(n,f)]}{\sigma[^{233}\mathrm{U}(n,f)]}$ and to benchmark the surrogate ratio technique for actinide nuclei. The data were taken using the STARS spectrometer operated in conjunction with the Liberace clover germanium detector array (the gamma-ray data were not used in this particular analysis). The alpha particle beam energy was chosen to be 55 MeV and

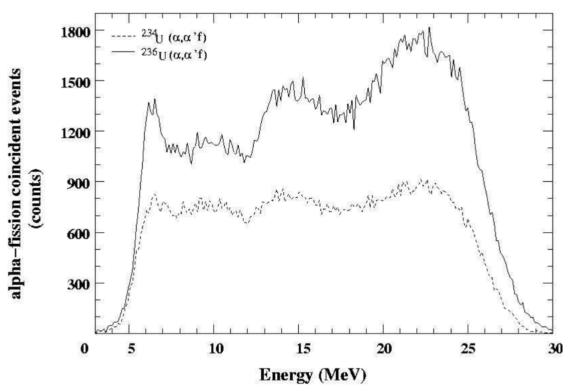

Fig. 2. Number of fission events for $^{234}\mathrm{U}(\alpha, \alpha' f)$ (top) and $^{236}\mathrm{U}(\alpha, \alpha' f)$ (bottom) plotted as a function of the excitation energy of the nucleus.

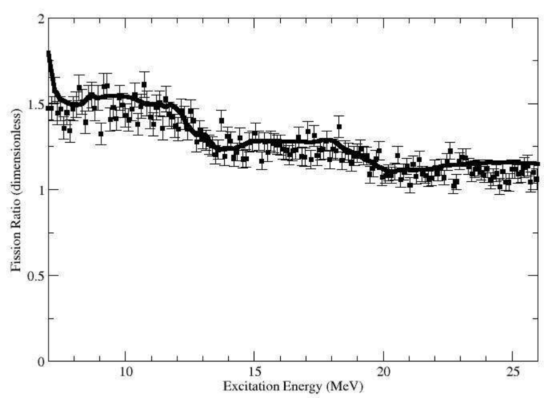

Fig. 3. Benchmarking the surrogate ratio method. The figure shows the cross section ratio for $\frac{\sigma[^{234}\mathrm{U}](n,f)}{\sigma[^{236}\mathrm{U}](n,f)}$ deduced using the surrogate ratio method (filled squares with error bars) compared to the directly measured ratio extracted from ENDF-87.

the beam was delivered by the 88-inch Cyclotron at the Lawrence Berkeley National Laboratory. The master trigger for the data acquisition required a signal in both the ΔE and E detectors.

Fig. 2 shows the number of fission events for each target plotted as a function of excitation energy, given by the beam energy minus the total energy of the scattered alpha particle. The total alpha particle energy is obtained from the sum of the energy collected in the ΔE and E detectors corrected for the energy losses in the delta-shield and dead layers of the detectors. The data in Fig. 2 has further been corrected for the different beam fluxes and target thicknesses for the two targets. As expected for low excitation energy (corresponding to high energies for the scattered alpha particle) there is little probability for fission. However, with increasing excitation energy, the fission probability at first rises rapidly before leveling off and then increasing in a series of steps corresponding to second and third chance fission.

Fig. 3 shows the ratio of fission probabilities $\frac{N(^{234}\mathrm{U}(\alpha,\alpha' f))}{N(^{236}\mathrm{U}(\alpha,\alpha' f))}$ (essentially the ratio of the two curves in Fig. 2), now plotted as a function of excitation energy. In Fig. 3 the data, indicated by the closed squares with error bars, are compared to the ratio of the directly-measured cross sections, $\frac{\sigma[^{233}\mathrm{U}(n,f)]}{\sigma[^{235}\mathrm{U}(n,f)]}$, as obtained from ENDF-87. As can be seen the agreement is excellent. The surrogate ratio is typically within 5% of the accepted value over an excitation energy range of $\sim 18\,\mathrm{MeV}$.

3. New results on conversion coefficients in TSD ^{167}Lu

In the second part of this paper new results for internal conversion coefficients of transitions depopulating triaxial strongly deformed bands in ^{167}Lu will be discussed.

The search for stable triaxial-deformed nuclei, rather than gamma-soft nuclei, has been ongoing for many years. Despite many experiments and much theoretical effort a unique signature for stable triaxial deformation has until very recently proved elusive. Recently, however, evidence has been found of the long predicted wobbling mode [15], which is a definitive signature of a stable triaxial nuclear shape. The experimental evidence consists of pairs of triaxial strongly deformed bands (TSD) in ^{163}Lu, ^{165}Lu, and ^{167}Lu, which show many of the characteristics expected of the wobbling mode [16–20] — see partial level scheme in Fig. 4.

In these band pairs, the excited TSD band decays primarily into the lower-lying band by a set of $\Delta I = 1$ non-stretched E2 transitions which connect states of spin $I \to I-1$. A partial level scheme of ^{167}Lu, shown in Fig. 4, shows the relevant pair of bands. As can be seen, the excited band decays into the lower lying band via a series of six $\Delta I = 1$ transitions, each with energy of about 700 keV. The assignment of the excited band as

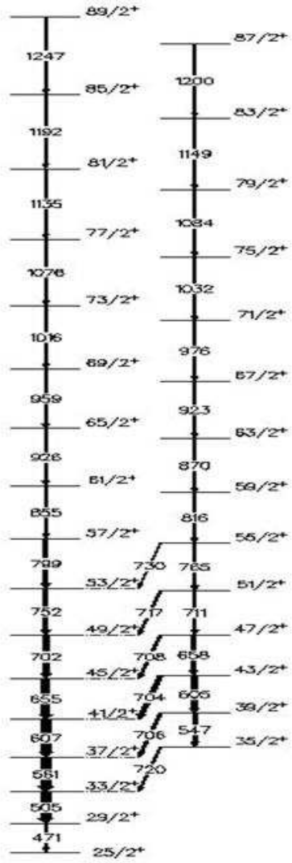

Fig. 4. Partial level scheme for ^{167}Lu showing the two strongest TSD bands. Band 2 (right) is a candidate for the wobbling mode [19]. The linking transitions have been assigned E2 character.

a wobbling phonon excitation based on the ground state TSD band follows from this decay pattern, from the mixed $\Delta I = 1$ M1/E2 character, with a strong E2 component, of the linking transitions and from the measured in-band/out-of-band branching ratios.

The relative spin assignments for the ^{167}Lu bands and the characterization of the linking transitions as mixed M1/E2 follow from angular-correlation measurements, which are difficult to carry out and sometimes subject to ambiguities. Our experiment aimed to confirm the M1/E2 character of the linking transitions between the wobbling band and ground state TSD band in ^{167}Lu by measuring their conversion coefficients, thus providing a direct and unambiguous measurement of the electromagnetic character of these transitions.

Gammasphere (consisting of 101 Compton suppressed Ge detectors for this experiment) coupled to the ICE Ball spectrometer was used to measure the internal conversion coefficients. The ICE Ball spectrometer [21] is a Gammasphere auxiliary detector that consists of six mini-orange electron spectrometers. Each mini-orange spectrometer had a ~5 mm thick SiLi detector to measure the electron energies. The strength and arrangement of the permanent magnets ensured that transmission efficiencies of four of the mini-orange spectrometers were optimized for 600–900 keV electrons. The transmission efficiencies of the remaining two detectors were optimized for lower energy electrons. The reaction chosen was ^{48}Ca + ^{123}Sb at a beam energy of 203 MeV, which populated high spin states in ^{167}Lu via the $4n$ channel. The self supporting ^{123}Sb target was $\sim 1.2\,\mathrm{mg/cm^2}$ thick. The ^{48}Ca beam was provided by the ATLAS accelerator at Argonne National Laboratory. The master trigger for the data acquisition required either five or more suppressed Ge detectors or at least one SiLi detector in ICE Ball plus at least two suppressed Ge detectors. Over a ten day period a total of $\sim 2\times 10^9$ events were recorded, the statistics being about equally split between pure gamma and electron-gamma coincidences. During the course of the experiment technical difficulties resulted in only four mini-orange channels working correctly. The in-beam electron resolution was $\sim$20 keV. It was dominated by Doppler broadening effects due to the high recoil velocity and the large opening angle of the mini-orange spectrometers (the resolution obtained with a calibration electron source is on the order of 2.0–3.0 keV at 1000 keV). More details of the experimental setup can be found in [22].

Figs. 5 and 6 show preliminary double-gated gamma and electron spectra for TSD band 1 (these spectra were obtained using only a single mini-orange spectrometer). As can be seen the quality of the spectra is high. Conversion electron peaks for several of the TSD band transitions are clearly visible, as are those for the lower-lying normal deformed transitions.

Using these data conversion coefficients were measured for seven in-band transitions of TSD Band 1 (from the 561 keV transition up to the 885 keV band member). These are shown in Fig. 7. All measured conversion coefficients are consistent with the expected E2 character of these rotational band members.

TSD Band 2 is considerably weaker than band 1. In addition, many of its transitions are close doublets with much more intense transitions elsewhere in the level scheme, or in TSD Band 1. Therefore, it has not yet been possible to extract similar spectra for TSD Band 2 or suitably gated spectra that emphasize the sought-after linking transitions. The data are still under analysis and we are confident that at the minimum a limit on the conversion coefficients for the linking transitions can be extracted from the data.

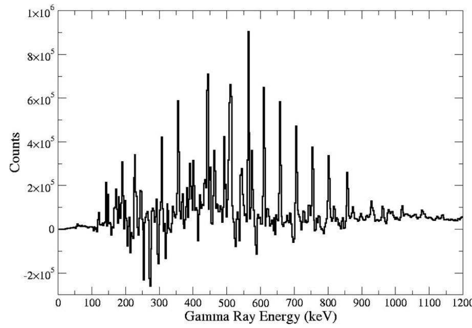

Fig. 5. Gamma–gamma gated gamma-ray coincidence spectrum for TSD Band 1 in ^{167}Lu. Transitions belonging to TSD band 1 are clearly visible as are transitions connecting lower-lying normal-deformed states. The spectrum is obtained by summing all pairs of gates on TSD Band 1 transitions from 505 to 1192 keV.

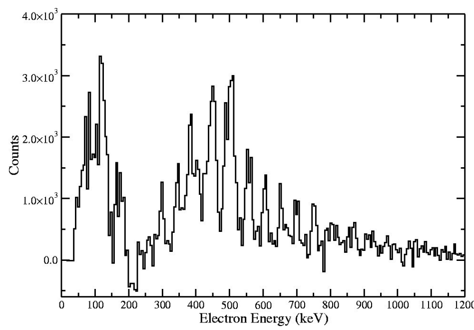

Fig. 6. Gamma–gamma gated electron spectrum for TSD Band 1. The exact same gating conditions were used as in Fig. 5. From $\sim$ 500 keV upwards, the peaks corresponds to (mostly) K-electron peaks from transitions in TSD Band 1.

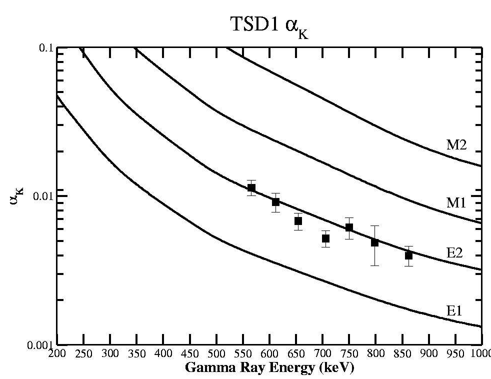

Fig. 7. Alpha-K conversion coefficients for TSD Band 1 in ^{167}Lu. The data are in excellent agreement with the expected E2 character for these transitions.

4. Conclusions

Two very disparate topics have been discussed in this paper. In the first section the surrogate ratio method was introduced and its effectiveness for deducing fission cross sections for unstable nuclei was discussed. New data for the cross section ratio $\frac{\sigma[^{233}\mathrm{U}(n,f)]}{\sigma[^{235}\mathrm{U}(n,f)]}$ was presented which indicates that under ideal circumstances the surrogate method may be accurate to $\sim 5\%$ over a wide range of equivalent neutron energies. In the second half of the paper new results for internal conversion coefficients of TSD bands in ^{167}Lu were presented.

These experiments would not have been possible without the help of the many superb professionals at the Wright Nuclear Structure Laboratory, the 88-inch Accelerator at Lawrence Berkeley National Laboratory and the ATLAS accelerator at Argonne National Laboratory. The invaluable assistance of these many individuals is gratefully acknowledged. The data analysis for the surrogate results is due to the hard work of C. Plettner, S.R. Lesher, J. Burke, L.A. Bernstein and H. Ai whose thesis work this represents. The ^{167}Lu experiment would not have been possible without the invaluable assistance of C.J. Lister and J. Roher, who together "saved" ICE Ball after it literally fell off a truck. The data analysis is primarily the work of G. Gurdal whose thesis work this represents.

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