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# To the continuum and beyond: Structure of U nuclei.

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**Abstract.** An experiment was performed at the 88-inch cyclotron at LBNL to investigate the structure of uranium isotopes and concurrently test the so-called surrogate ratio method. A 28 MeV proton beam was used to bombard $^{236}$U and $^{238}$U targets and the outgoing light ions were detected using the STARS silicon telescope allowing isotopic assignments and the excitation energy of the compound nucleus to be measured. A fission detector was placed at backward angles to give particle-fission coincidences, while the six clover germanium detectors of the LIBERACE array were used for particle-$\gamma$ coincidences. The (p,d) reaction channels on $^{236}$U and $^{238}$U targets were used as a surrogate to measure the $\sigma(^{234}\mathrm{U(n,f)})/\sigma(^{236}\mathrm{U(n,f)})$ cross section ratio. The results give reasonable agreement with literature values over an equivalent neutron energy range between 0 MeV and 6 MeV. Structure results in $^{235}$U include a new $(3/2^-)$ level at 1035 keV, that is tentatively assigned as the $3/2^-[501]$ Nilsson state. The analogue $3/2^-[501]$ state in $^{237}$U may be associated with a previously observed level at 1201 keV, whose spin/parity is restricted to $J^\pi = 3/2^-$ on the basis of newly observed decays to the ground band.

## 1. Introduction

The STARS array (Silicon Telescope Array for Reaction Studies) in conjunction with the LIBERACE array (LIvermore BERkeley Array for Collaborative Experiments) at the 88-Inch Cyclotron at LBNL is proving to be an extremely powerful device to study nuclear structure via (charged) particle-$\gamma$ coincidence spectroscopy [1]. The technique enables the properties of low- and medium-spin states up to and beyond the neutron separation energy to be probed and gives a rare insight into the $\gamma$-ray continuum. In our experiments the nuclei of interest are generally populated via light-ion charged particle transfer reactions. The exit channel of interest and excitation energy of the residual nucleus are selected by measuring the outgoing charged particle using the segmented Si detectors of the STARS array, while coincident gamma rays are detected with the LIBERACE Clover array. The resulting coincident particle spectrum shows the ensemble of states that were directly populated by the reaction while gamma-ray coincidences reveal the decay path from a given level. This sensitivity extends to very high excitation energies in the continuum, allowing for correlations between low-lying discrete levels (both collective and single particle) and, for example, the spin distribution of the entry states in the continuum region.

Over the last few years, our research using STARS/LIBERACE has focused on testing and utilizing the so-called surrogate method. This method was first employed in 1970 [2], and has recently been redeveloped to emerge as an effective means of indirectly measuring neutron-induced fission cross sections [3-11]. Surrogate reactions use a stable beam and target combination to populate the same compound nucleus as that formed in a neutron-induced reaction of interest. The fission probability of the compound nucleus is then measured in the surrogate experiment in order to extract $\sigma$(n,f).

A recent experiment using (p,d) and (p,t) reactions on $^{236}$U and $^{238}$U targets was performed with two main aims: (1) To test the surrogate technique, and specifically the surrogate ratio method, using (p,d) reactions as a surrogate to measure the $\sigma(^{236}\text{U(n,f)})/\sigma(^{234}\text{U(n,f)})$ cross section ratio. (2) Utilisng particle-$\gamma$ coincidence spectroscopy to study specific states in Uranium isotopes selectively populated in these light ion reactions.

## 2. Experimental Procedure

A 28 MeV proton beam from the 88" Cyclotron was used to bombard $^{236}$U and $^{238}$U targets at Lawrence Berkeley National Laboratory. For this experiment STARS was configured as a $\Delta$E-E particle telescope consisting of three S2 type silicon detectors - each segmented into 8 sectors and 24 concentric 1 mm rings - which was used to measure the energy and angles of outgoing light ions within an angular range of $34^o$-$64^o$. Another Micron S2 detector was placed upstream of the target for fission fragment detection and covered angles between $109^o$ and $137^o$. A 4 mg Al $\Delta e^-$ shield was placed between the target and the $\Delta$E detector, both to stop forward going fission fragments from entering the $\Delta$E detector and to help reduce the effect of $\delta$-electrons produced in the target. The STARS chamber was surrounded by the six clover detectors of the LIBERACE array enabling $\gamma$-ray detection. The STARS/LIBERACE setup is shown schematically in figure 1.

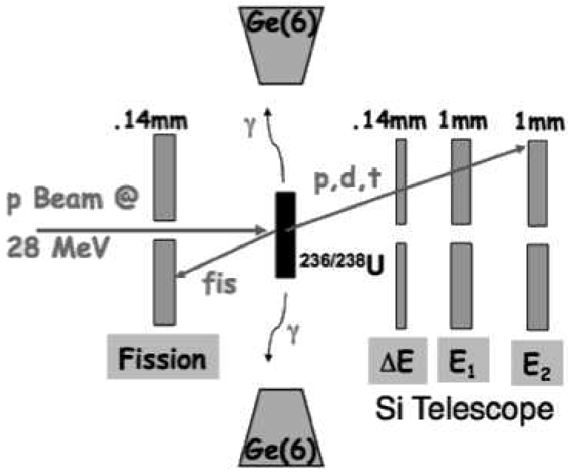


**Figure 1.** Schematic diagram of the STARS/LIBERACE array at LBNL.

The $^{236}$U and $^{238}$U targets had thicknesses of 322(18) $\mu$g/cm$^2$ and 1450(100) $\mu$g/cm$^2$, respectively. For both targets, contaminant species were less than 1%. Data were collected for 25 hours on the $^{236}$U target with an average proton beam intensity of 1.04 enA and an average 90.7 % live time. For the $^{238}$U target, the integrated beam time was 70 hours with an average intensity of 0.70 enA and an average live time of 88.1 %.

## 3. Benchmarking the (p,df) surrogate reaction

According to the Hauser-Feshbach formalism [12], the total cross section of a neutron-induced fission reaction is

$$\sigma_{(n,f)} = \sigma_n^{CN}(E^*, J, \pi)\ G_{(n,f)}^{CN}(E^*, J, \pi) \tag{1}$$

where $\sigma_n^{CN}$ defines the cross section of compound nucleus formation in the neutron reaction and $G_{(n,f)}^{CN}$ defines the probability that the compound nucleus will decay by fission. Both terms depend on the excitation energy, spin and parity of the compound nucleus. In the Weisskopf-Ewing limit [13], the fission decay probability, $G_{(n,f)}^{CN}$, is assumed to depend only on the excitation energy (i.e., to be independent of the spin and parity). The neutron-induced formation cross section, $\sigma_n^{CN}$, can typically be calculated using an optical model, whereas $G_{(n,f)}^{CN}(E^*)$ must be directly measured.

If the desired neutron induced fission reaction is written as A + n → C*, where C* is a compound nucleus state, then the surrogate reaction to populate the same compound nucleus is given by B + b → C* + c, and the fission decay probability, $G_{(n,f)}^{CN}(E^*)$, can be measured directly via the surrogate reaction, $G_{(b,cf)}^{CN}$, according to

$$G_{(n,f)}^{CN}(E^*) = G_{(b,cf)}^{CN}(E^*) = \frac{N_{(b,cf)}}{\epsilon_f N_{(b,c)}}, \tag{2}$$

in which $\epsilon_f$ is the fission detection efficiency, $N_{(b,cf)}$ is the number of fission events detected in coincidence with the surrogate reaction and $N_{(b,c)}$ is the total number of compound nuclei, C* produced. Determination of $N_{(b,c)}$ can be very difficult due to target impurities, and the requirement of carbon or metal backings for some actinide targets. Because of this, the surrogate ratio technique was developed and is discussed below.

In the surrogate ratio method [4, 14], two surrogate measurements with identical reactions and setup are performed on neighbouring nuclei with very similar structure. The ratio of the fission decay probabilities from the two measurements can then be taken, thus eliminating $N_{(b,c)}$ and $\epsilon_f$ in equation 2. It is assumed that the neutron induced compound nucleus formation cross sections for the two reactions also cancel, so that the fission probability ratio of two surrogate experiments, x and y, can be used to directly determine the ratios of two (n,f) cross sections according to

$$\frac{\sigma_{(n,f)}^x}{\sigma_{(n,f)}^y} = C\frac{N_{(b,cf)}^x(E^*)}{N_{(b,cf)}^y(E^*)}, \tag{3}$$

in which the normalization constant C is independent of energy and is given by

$$C = \frac{(\rho_T \ell_t Q)^y}{(\rho_T \ell_t Q)^x}, \tag{4}$$

where the terms represent areal target density ($\rho_T$), experimental live time ($\ell_t$), and integrated charge delivered by the particle beam ($Q$).

The particle spectra for $^{238}$U(p,df)$^{237}$U and $^{236}$U(p,df)$^{235}$U are presented in figure 2. The $^{236}$U(p,df) spectrum has been normalised to the $^{238}$U(p,df) data according to equations 3 and 4, and both spectra have been compressed to 100 keV/chan. The energy spectra in figure 2 are adjusted so that the equivalent neutron energy, $E_n$, is zero at the approximate neutron separation energies for $^{235}$U and $^{237}$U (Sn≈5.1 MeV for both [15], [16]). There is a distortion beyond $E_n \sim 6$ MeV in both spectra. This energy corresponds to deuteron energies of ∼ 12 MeV (and below), and can be ascribed to the effect of the Coulomb barrier ($V_c$) that lies at ≈12 MeV for the $^{235}$U-d and $^{237}$U-d systems.

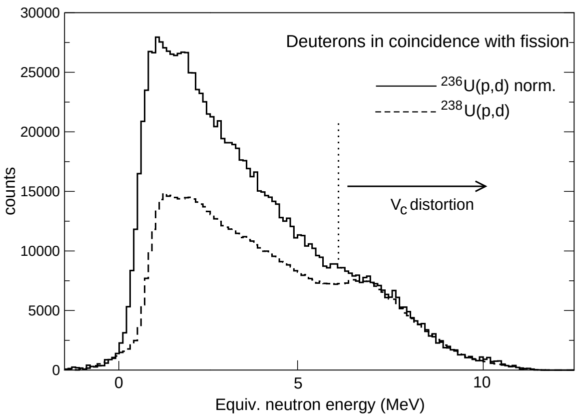


**Figure 2.** Comparison of deuteron spectra for the $^{238}$U(p,d)$^{237}$U and $^{236}$U(p,d)$^{235}$U reactions. The $^{236}$U(p,d)$^{235}$U data have been normalised to correct for the different integrated beam and target thicknesses in the two experiments. The region beyond ~6 MeV is distorted as described in the text.

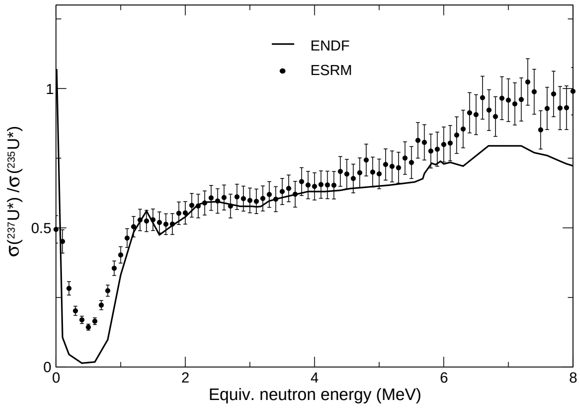


**Figure 3.** $\sigma(^{236}\text{U(n,f)})/\sigma(^{234}\text{U(n,f)})$ ratios measured by the $^{238}$U(p,d)$^{237}$U/$^{236}$U(p,d)$^{235}$U surrogate measurement (data points) and compared to the literature value for the ratio (solid line).

The $\sigma(^{236}\text{U(n,f)})/\sigma(^{234}\text{U(n,f)})$ cross section ratio extracted from the $^{238}$U(p,df))/$^{236}$U(p,df) experimental surrogate ratio (points with error bars) is compared to the ratio from the ENDF VII literature values (solid line) in figure 3. The data show reasonably good agreement up to $E_n \approx 6$ MeV. Below 1 MeV the surrogate ratio does not reproduce the large difference in the two cross section precisely, although the trend is reproduced. Beyond equivalent neutron energies of $E_n \sim 6$ MeV, there is a rapid deviation of the surrogate measurement from the directly measured cross section ratio due to the outgoing deuterons having energies below $V_c \approx 12$ MeV, as discussed above.

## 4. Single particle states in $^{235}$U and $^{237}$U

As discussed in Section I., STARS/LIBERACE allows particle-$\gamma$ coincidence measurements to be used for structure studies of selected states populated in excited nuclei via light ion reactions. Figure 4 (a) shows the deuteron energy spectrum in coincidence with the 646 keV $\gamma$ ray that deexcites the $1/2^-[501]+1/2^-[770]$ state in $^{235}$U. In addition to weak continuum population, the spectrum indicates strong direct population of the 659 keV level and also direct population of a ~1000 keV level that subsequently decays to the 659 keV level. Figure 4 (b) shows $\gamma$ rays in coincidence with deuterons associated with the higher energy peak in figure 4 (a). In addition to the expected 646 keV and 659 keV transitions (which depopulate the known $1/2^-$ state at 659 keV), the spectrum shows a number of new $\gamma$ rays (marked with arrows). Further analysis of these $\gamma$ rays leads to the partial decay scheme for $^{235}$U shown in figure 6.

Turning to $^{237}$U, the deuteron spectrum observed in coincidence with the 854 keV $\gamma$ ray decay from the $1/2^-[501]$ state in $^{237}$U is shown in figure 5 (a). As for the analogous state in $^{235}$U, there is strong direct population of the $1/2^-[501]$, 865 keV level and a smaller peak at higher energy corresponding to a higher-lying level. Figure 5 (b) shows a $\gamma$-ray spectrum requiring coincidences with the 1200 keV deuteron peak in figure 5 (a). The level scheme for $^{237}$U has been extended to include new $\gamma$ rays seen in figure 5 (b) (marked with arrows) and is shown in figure 6.

The partial decay schemes for $^{235}$U and $^{237}$U shown in figure 6 highlight the very similar low-spin structure in these neighbouring odd-mass nuclei. As has been previously observed for both $^{235}$U [17] and $^{237}$U [18], pickup reactions exhibit strong population of the $1/2^-[501]$ Nilsson

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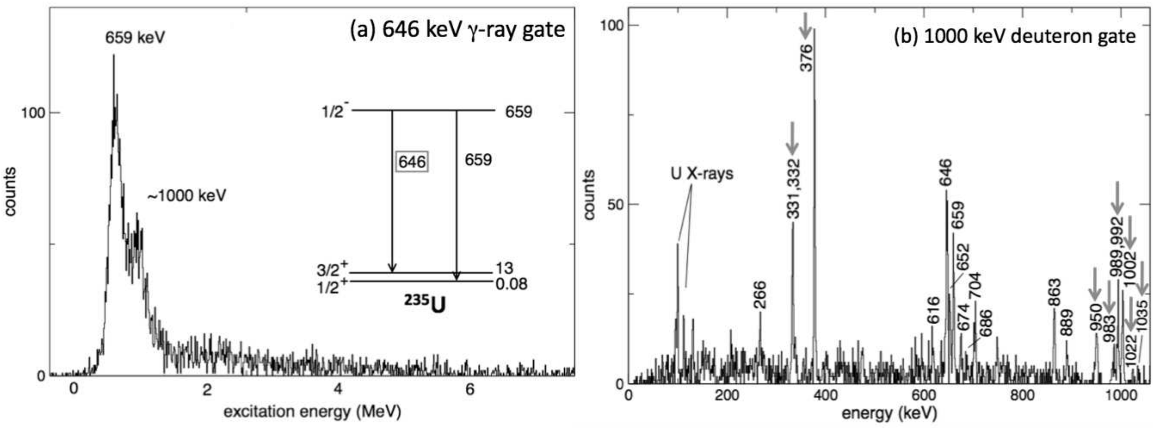


**Figure 4.** Spectra for $^{235}$U. (a) Deuterons in coincidence with the 646 keV $\gamma$ ray. (b) $\gamma$ rays in coincidence with the 1000 keV peak from (a). $\gamma$ rays of interest are marked with arrows.

level (for $^{235}$U, the 659 keV level is believed to be a $1/2^-[501]+1/2^-[770]$ mixed state [17]). This large cross section is partially due to the level being close to and below the Fermi energy, and also due to the overlap between the angular momentum transfer in the (p,d) reaction (L=1) and the spin of the spherical parent of the $1/2^-[501]$ state ($3p_{1/2}$).

While the $1/2^-[501]$ Nilsson state has been assigned in both $^{235}$U and $^{237}$U, the $3/2^-[501]$ state remains unobserved. Nevertheless, this level should also be populated with relatively large cross section, as it also lies just below the Fermi surface, and displays a large overlap between the angular momentum transfer, L=1, and the spin of the spherical parent ($3p_{3/2}$). In the case of $^{235}$U, a 1002 keV level was previously observed and given a tentative ($1/2^-$,$3/2^-$) spin/parity assignment. While this level is also observed weakly in the present work, a new 1035 keV state is measured with higher cross section as indicated by strong 331 keV and 376 keV transitions to the $1/2^-$ and $3/2^-$ members of the $1/2^-[501]$ band (see figure 4 (b)). This level has $J^\pi = (3/2^-)$ based on observed decays to states with known spin and parity (in particular the positive parity, $1/2^+$ band at low energy), and is tentatively assigned as the expected $3/2^-[501]$ state. For $^{237}$U, figures 5 (a) and (b) imply strong direct population of the level at 1201 keV. This state was

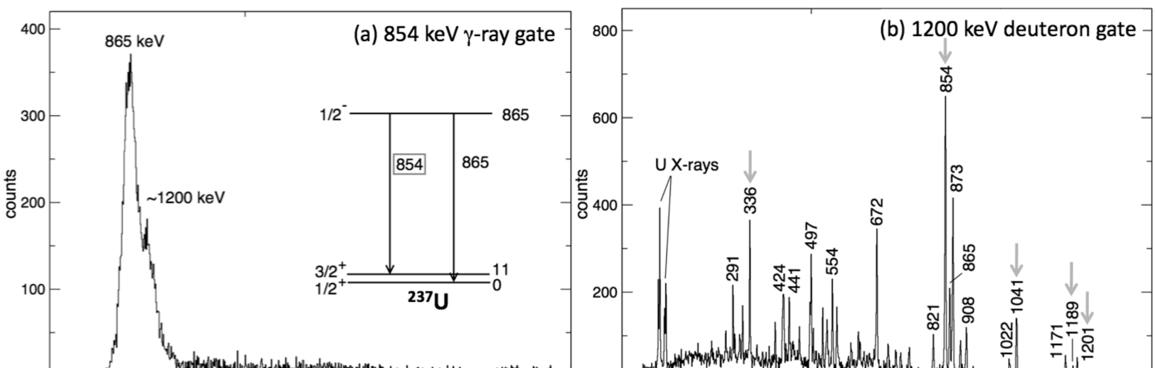


**Figure 5.** Spectra for $^{237}$U. (a) Deuterons in coincidence with the 854 keV $\gamma$ ray. (b) $\gamma$ rays in coincidence with the 1200 keV peak from (a). $\gamma$ rays of interest are marked with arrows.

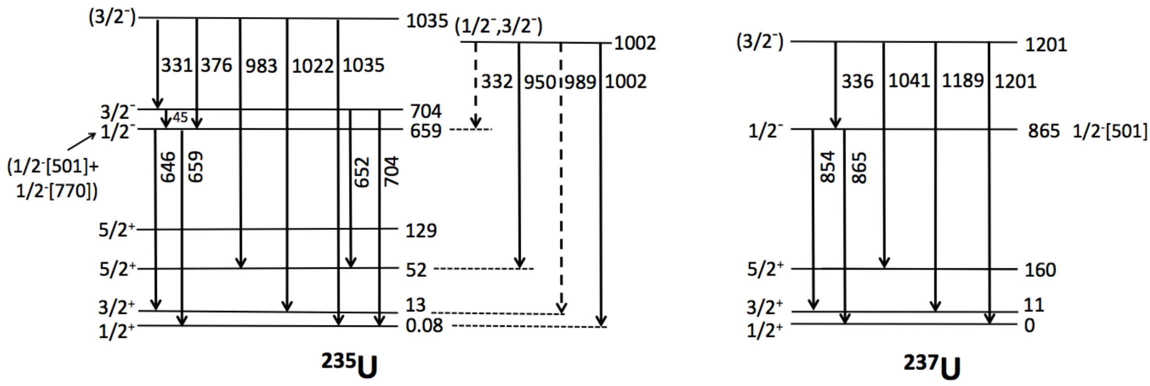


**Figure 6.** Partial decay schemes for $^{235}$U and $^{237}$U.

previously observed and assigned a spin and parity $J^\pi = (1/2^-, 3/2^-)$. Newly observed decays including a 1041 keV decay to the $5/2^+$ state at 160 keV limit the spin/parity to $J^\pi = 3/2^-$. The 1201 keV level is therefore a good candidate for the $3/2^-[501]$ Nilsson state in this nucleus.

In conclusion the low/medium spin structure of $^{235}$U and $^{237}$U have been probed via charged particle-induced transfer reactions. Particle-$\gamma$ coincidences provide a powerful tool for spin and level assignments. Results include the possible identification of the $3/2^-[501]$ intrinsic Nilsson state at energies of 1035 keV and 1201 keV in $^{235}$U and $^{237}$U, respectively. In parallel with these structure results, the efficacy of the surrogate ratio method has been probed for (p,d) reactions in the actinides. Reasonable agreement is found between the $\sigma(^{236}\mathrm{U(n,f)})/\sigma(^{234}\mathrm{U(n,f)})$ cross section ratio extracted from the present surrogate experiment and accepted direct neutron induced cross section results up to about 6 MeV equivalent neutron energy (~11 MeV excitation energy).

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