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# Neutron Capture on Actinides studied with DANCE

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**Abstract.** Neutron capture cross sections in the continuum region above about neutron energies of 1 keV have been difficult to calculate accurately, and measurements are needed for accurate results. Recent capture measurements on  $^{234,236,238}U$  and  $^{239}Pu$  made using the DANCE detector have resolved some discrepancies in the  $^{235}U$  and  $^{239}Pu$  capture cross sections. In addition, measurements of the gamma-ray emission spectra from capture are shown to provide an additional constraint on the photon strength functions used in calculating the spectrum shape and cross section. Using photon strength functions constrained by the gamma-ray spectra, cross-section calculations can be made that reproduce the measurements with no additional renormalization.

**Keywords:** neutron capture, photon strength function, uranium, plutonium, actinide

## 1 Introduction

Neutron capture cross sections in the continuum region have been difficult to calculate accurately [1], and measured cross sections are favored when accurate results are needed. Calculations usually use the Hauser-Feshbach approach (eq. 1), where  $k_n$  is the neutron wave number,  $g_c$  is a statistical spin factor,  $T_n$  is the neutron transmission coefficient,  $T_\gamma$  is the  $\gamma$ -ray transmission coefficient, and  $W_{n\gamma}$  is the width fluctuation factor. Good agreement with measurements is often obtained if  $T_\gamma$  is normalized to the measured s-wave resonance spacing  $D_0$  and average s-wave radiation width  $\langle \Gamma_\gamma \rangle$ ,  $T_\gamma = 2\pi \langle \Gamma_\gamma \rangle / D_0$ .

$$\sigma_{capt}(E_n) = \frac{\pi}{k_n^2} \sum_{J\pi} g_c \frac{T_n T_\gamma}{T_n + T_\gamma} W_{n\gamma}, \quad (1)$$

The gamma-ray transmission coefficient is defined as

$$T_\gamma = \sum_{j^\pi XL} \int_0^{E'} 2\pi E_\gamma^{(2L+1)} f_{XL}(E_\gamma) \rho(E_x, j^\pi) dE_x \quad (2)$$

where  $f_{XL}(E_\gamma)$  is the photon strength function and  $\rho(E_x, j^\pi)$  is the nuclear level density.

There has been a great deal of recent progress in understanding capture calculations, including studying the low-energy behavior of the E1 strength function, the Oslo method for determining strength functions and level densities, the recognition of the need for additional components in the strength function in addition to the E1 GDR, and QRPA extensive calculations by Goriely, Hilaire, and co-workers [2].

In addition to the capture cross section, the shape of the gamma-ray cascade spectrum can be measured and compared to calculations, providing an additional constraint on the strength function and level density. Our plan is to measure the  $\gamma$ -ray spectra from discrete neutron capture resonances using DANCE and compare to calculations made using the DICEBOX code [3] propagated through a GEANT-4 model of DANCE [4]. We will then vary the strength function models and parameters to achieve a "good" description of the measured spectra. The "best" parameters will then be used to calculate the capture cross section at  $E_n \geq 2$  keV using the CoH<sub>3</sub> Hauser-Feshbach code [5].

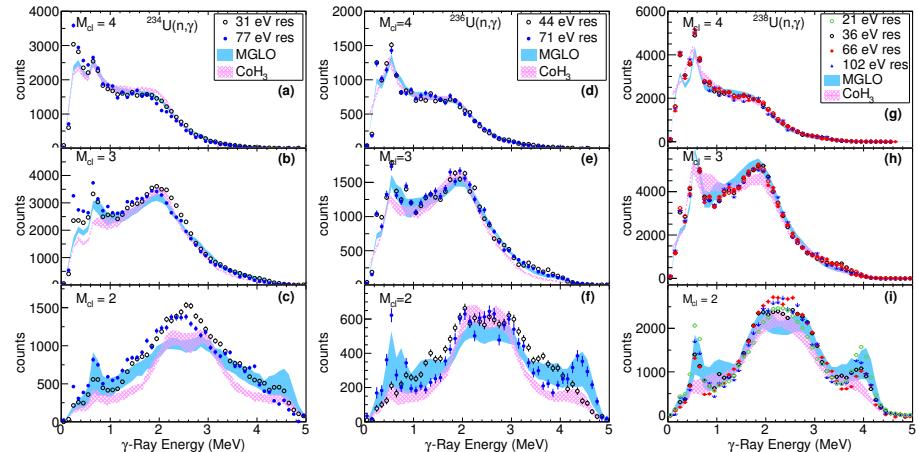
## 2 DANCE

The Detector for Advanced Neutron Capture Experiments (DANCE) is a nearly  $4\pi$  BaF<sub>2</sub> array consisting of 160 crystals of 4 different shapes, each with a volume of 734 cm<sup>3</sup>. It is highly efficient so that resonance-region measurements can be made with less than 1 mg/cm<sup>2</sup> of material, but good results for neutron energies greater than 1 keV require somewhat thicker samples. A <sup>6</sup>LiH sphere surrounds the target location to attenuate scattered neutrons. The detector is located on FP-14 at the Los Alamos Neutron Science Center, 20.25 m from the upper-tier water moderator. DANCE is a calorimetric detector, capable of detecting and summing the complete gamma cascade following capture. If all of the cascade gammas are detected, the summed energy is the Q value of the capture reaction, and can be used to identify capture events.

## 3 Results for even Uranium isotopes

The gamma-ray spectra from neutron capture on <sup>234,236,238</sup>U were studied using targets from 1 to 3 mg/cm<sup>2</sup> thick. For these isotopes, the continuum fission cross section is negligible below about 0.5 MeV. DICEBOX calculations of the spectra were made using the prescription of Kopecky and Uhl [6] for the photon strength function: a generalized Lorentzian form (GLO) for the E1 giant dipole, and standard Lorentzians (SLO) for the M1 "spin flip" resonance and

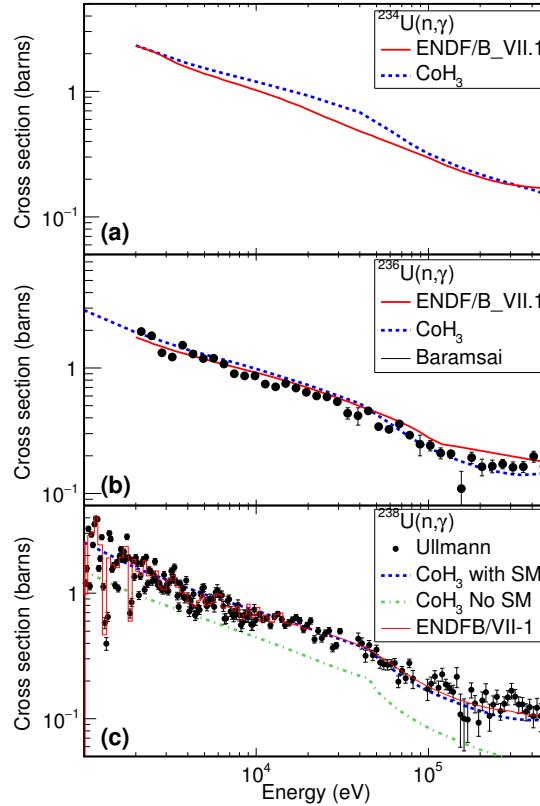
E2 contribution. The parameters for the giant dipole were taken from the Empire compilation [10], and for the spin-flip and E2 contributions from RIPL-3. In addition, a low-lying (about 2-3 MeV) M1 component, consistent with the scissors mode resonance, was also included. The scissors mode was represented by 2 standard Lorentzians, at 2.15 and 2.90 MeV. The energy and width of the Lorentzians were taken from an Oslo-method analysis [7], while the strengths were varied to provide the best representation of the spectra. The same parameters were used for all three isotopes. It was shown in ref. [8] that the gamma ray spectra could not be reproduced by the Kopecky and Uhl presecription, but additional strength at low energies, most likely M1, was required. That analysis was updated and extended to  $^{234,236}U$  in ref. [14], with results shown below in fig. 1. Fig. 1 shows gamma-ray spectra for several  $1/2^+$  resonances in each isotope and for several gamma-ray multiplicities. The calculations labelled CoH<sub>3</sub> were made using the generalized Lorentzian form for the giant dipole with parameters from ref [10]. The calculations labelled MGLO were made using the modified generalized Lorentzian form [11] for the giant dipole with parameters from ref. [12].



**Fig. 1.** Measured  $\gamma$ -ray spectra for several  $1/2^+$  resonances in  $^{234,236,238}U(n,\gamma)$  compared to calculations made with photon strength-function and nuclear level-density parameters obtained from systematics used in the CoH<sub>3</sub> code and using the MGLO and CoH<sub>3</sub> models for the E1 strength. The resonance energies are indicated in each panel. The y-axis counts are arbitrarily normalized.

Figure 2 shows the cross sections calculated with CoH<sub>3</sub> compared to measurements. The calculations used the GLO form of the photon strength function with parameters described above. The data for  $^{238}U$  is from ref. [8], and was normalized to low-lying resonances. The data for  $^{236}U$  is from ref. [13] and was normalized with experimentally-determined efficiencies. There is no data for  $^{234}U$

in the EXFOR data base; a recent measurement at DANCE will be analyzed. The  $^{234}\text{U}$  calculation is compared to the ENDF/B-VII.1 evaluation. Note the calculations are absolute and not renormalized to the data; very good agreement in magnitude and shape was obtained.

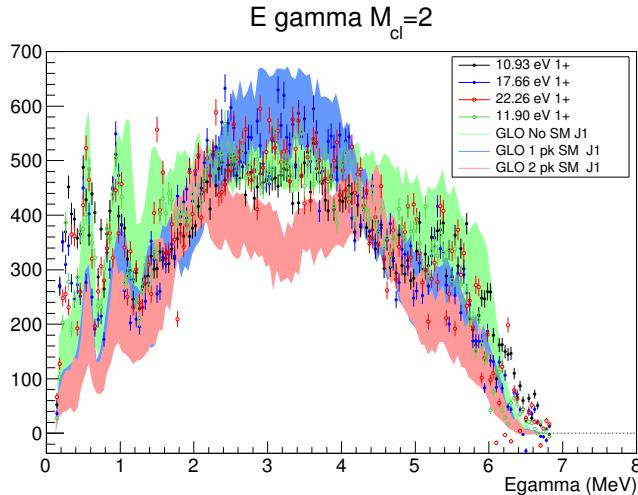


**Fig. 2.** Uranium capture cross sections compared to calculations

#### 4 Results for $^{239}\text{Pu}$

Measurements of neutron capture on  $^{239}\text{Pu}$  are complicated by the large fission cross section for  $^{239}\text{Pu}$ , and fission tagging is crucial. The gamma-ray spectrum following capture was measured with a  $2.43 \text{ mg/cm}^2$   $^{239}\text{Pu}$  target mounted in a

parallel-plate avalanche counter inserted at the target location of DANCE [15]. A preliminary analysis of the multiplicity-two spectrum from several resonances is shown in fig. 3. DICEBOX calculations of the cascade were made using the GLO form for the giant dipole, with parameters taken as for the U isotopes. However, a two-Lorentzian scissors mode did not provide satisfactory results, and a one-Lorentzian scissors mode with parameters from the global systematics of ref. [1] was used.



**Fig. 3.** Measured  $\gamma$ -ray spectra for several  $1^+$  resonances in  $^{239}\text{Pu}(n, \gamma)$  compared to calculations made with photon strength-function and nuclear level-density parameters described in the text, and using the GLO models for the E1 GDR strength. The y-axis counts are arbitrarily normalized.

An accurate measurement of  $^{239}\text{Pu}(n, \gamma)$  from 1 keV to 1 MeV, made at DANCE, was recently reported by Mosby, et al.[16]. This measurement used a 50 mg Pu target, with fission corrections made based on the thin-target PPAC data. Cross section calculations were made using the CoH<sub>3</sub> code with parameters similar to those used in the gamma spectrum calculation, and a very good agreement with data was obtained, without renormalization.

## 5 Summary

We have shown that gamma-cascade spectra provide another test and constraint on the strength function and level densities used in capture cross section calculations. The standard Kopecky-Uhl prescription, consisting of a GLO giant-dipole form plus a standard Lorentzian "M1 spin-flip" and E2 contribution is not sufficient to calculate the shape of gamma-ray spectra observed in  $^{234,236,238}\text{U}(n, \gamma)$

and  $^{239}Pu(n,\gamma)$  reactions, and that additional strength at low energies (2 to 3 MeV), most likely an M1 scissors-mode resonance, is required. Accurate calculations of the capture cross section can be made by including the scissors mode in the photon strength function, with proper choice of models for the level density and E1 portion of the strength function. We note that the calculations are sensitive to the input parameters, and precise data and theory are needed to fix the models and their parameters.

## References

1. Mumpower, M.R. et al.: Estimation of M1 scissors mode strength for deformed nuclei in the medium to heavy mass region by statistical Hauser-Feshbach model calculations. *Phs. Rev. C* **96**, 024612 (2017).
2. See contribution by S. Hilaire, et al., to this conference.
3. Bečvář, F.: Simulation of  $\gamma$  cascades in complex nuclei with emphasis o assessment of uncertainties of cascade-related quantities. *Nucl. Instr. Meth. A* **417**, 434 (1998).
4. Jandel, M., et al.: GEANT4 simulations of the DANCE array. *Nucl. Instru. and Meth. B* **261**, 1117 (2007).
5. Kawano, T., Talou, P., Chadwick, M.B., and Watanabe,T.: Monte Carlo Simulation for Particle and  $\gamma$ -ray Emissions in Statistical Hauser-Feshbach model. *J. Nucl. Sci. Technol.* **47**, 462 (2010)
6. Kopecky, J. and Uhl, M.: Test of gamma-ray strength functions in nuclear reaction model calculations. *Phys. Rev. C* **41**, 1941 (1990).
7. Guttormsen, M. et al.: Scissors resonance in the quasicontinuum of Th, Pa, and U isotopes. *Phys. Rev. C* **89**, 014302 (2014).
8. Ullmann, J.L., et al.: Cross section and  $\gamma$ -ray spectra for  $^{238}U(n,\gamma)$  measured with the DANCE detector array at the Los Alamos Neutron Science Center. *Phs. Rev. C* **89**, 034603 (2014).
9. Ullmann, J.L., et al.: Constraining the calculation of  $^{234,236,238}U(n,\gamma)$  cross sections with measurements of the gamma-ray spectra at DANCE. *Phys. Rev. C* **96**, 024627 (2017).
10. Herman, M.: Technical report INDC(NDS)-0603, 2013 (unpublished.)
11. Kroll, J., et al.: Strength pf the scissors mode in odd-mass Gd isotopes from the radiative capture of resonance neutrons. *Phys. Rev. C* **88**, 034317 (2013).
12. Dietrich, S.S. and Berman, B.L.: Atlas of photoneutron cross sections obtained with monoenergetic photons. *Atomic Data and Nuclear Data Tables* **38**, 199 (1988).
13. Baramsai, B., et al.: Radiative neutron apture cross section from  $^{236}U$ . *Phs. Rev. C* **96**, 024619 (2017).
14. Ullmann, J.L., et al.: Constraining the calculation of  $^{234,236,238}U(n,\gamma)$  cross sections with measurements of the gamma-ray spectra at DANCE. *Phys. Rev. C* **96**, 024627 (2017).
15. Wu, C.-Y., et al.: A compact gas-filled avalanche counter for DANCE. *Nucl. Instrum. Meth. A* **694**, 78 (2012).
16. Mosby, S., et al.: Unifying measurement of  $^{239}Pu(n,\gamma)$  in the keV to MeV energy regime, *Phys. Rev. C* **97**, 04160R (2018).