

Scientists Use GEANIE to Study Isotopes of Iridium and Europium to Improve Radiochemical Diagnostics in Nuclear Devices

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Introduction

Radiochemical diagnostics play an important role in helping scientists understand the detonation of a nuclear device. Sometimes some elements or isotopes are inserted as radiochemical detectors at various locations in the nuclear device. During the detonation of the device, these detectors are subjected for a short time to the intense flux of neutrons emitted through fission and possibly through fusion of light elements (usually deuterium and tritium). After the detonation, the radiochemical detectors and their long-lived activation products are retrieved from the area where the underground explosion took place. These radiochemical samples are analyzed to extract information about how the device operated. A large amount of such radiochemical data exist from past nuclear-device tests.

Radiochemical detectors

Radiochemical detectors with an energy threshold in their activation cross section (i.e., the energy-dependent probability that incident neutrons produce the long-lived reaction product of interest in the detector) are used to measure a specific part of the neutron spectrum. For example, $(n,2n)$ reactions, which have thresholds in the range of 6 to 12 MeV, are mainly sensitive to fusion neutrons that are emitted with an energy of 14 MeV. These same reactions are relatively insensitive to fission neutrons that are emitted with an average energy of 2 MeV. Other reactions with different energy thresholds, such as neutron-inelastic scattering, can be used to measure other parts of the neutron-energy spectrum.

Radiochemical detectors are chosen to yield certain radioactive products that can be easily retrieved and have suitable lifetimes and decay schemes. From measurements of radioactivity in the debris, scientists can determine for each detector the quantity of long-lived radioactive products formed during the detonation. A basic ingredient in successfully using radiochemical detectors in the diagnostics of nuclear devices is an accurate knowledge of the activation cross sections of these detectors. The quantity of radioactive products produced depends on the activation cross section of the radiochemical detector and on the number and energies of the neutrons irradiating the detector. From the amount of radioactive products, scientists can deduce the neutron fluence, which is the flux integrated over time — a period that covers the entire duration of the detonation — of neutrons with sufficient energy to cause the formation of the radioactive product under investigation.

Because the neutron-energy spectrum and the location of the detector vary during the detonation of the nuclear device, the formation of reaction products from neutron irradiation of the detector is a time-dependent process. This process can only be determined from a simulation of the nuclear explosion using powerful computers. Although radiochemical detectors only provide macroscopic diagnostics, they put invaluable constraints on the simulation of nuclear detonations.

Los Alamos/Livermore Collaboration Uses the Unique Capabilities of GEANIE to Investigate Iridium and Europium Isotopes

Scientists from Lawrence Livermore National Laboratory (LLNL) and Los Alamos National Laboratory (LANL) are studying iridium and europium isotopes because of their use in radiochemical diagnostics. These isotopes differ in their nuclear properties and therefore present different problems and challenges in measuring their activation cross sections as a function of incident-neutron energy. These cross sections have been difficult to measure by conventional means and thus have not been accurately determined — until recently. The LANL/LLNL team has developed a more accurate method for determining the activation cross sections with the use of GEANIE (Germanium Array for Neutron-Induced Excitations), a large γ -ray detector array located at the Los Alamos Neutron Science Center's (LANSCE) Weapons Neutron Research Facility (WNR). GEANIE, which is equipped with planar detectors with excellent gamma-ray energy resolution (i.e., with a full-width-at-half-maximum resolution of 700 eV at 122 keV), is well suited to resolve the complex and specific γ -ray cascades that subsequently will help the scientists identify the radioactive products of interest. From the intensities of measured γ -ray cascades, the scientists can use Hauser-Feshbach nuclear-model calculations to calculate the γ -ray cascades not detected in the measurement and thus determine the needed activation cross sections. Similar measurements and calculations have already been carried out successfully for the $^{235}\text{U}(n,2n)^{234}\text{U}$ and $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ reactions, which are very difficult to measure.

Iridium Isotopes

Natural iridium consists of ^{191}Ir (37.3%) and ^{193}Ir (62.7%) — both of which can serve as radiochemical detectors. A long-lived state in ^{193}Ir (called $^{193\text{m}}\text{Ir}$, where “m” stands for “metastable”) at an excitation energy of 80.22 keV and with a lifetime of 10.53 days (see Fig. 1) can serve as a neutron-fluence detector most sensitive to low-energy neutrons (with energies above 80.22 keV) through inelastic scattering.

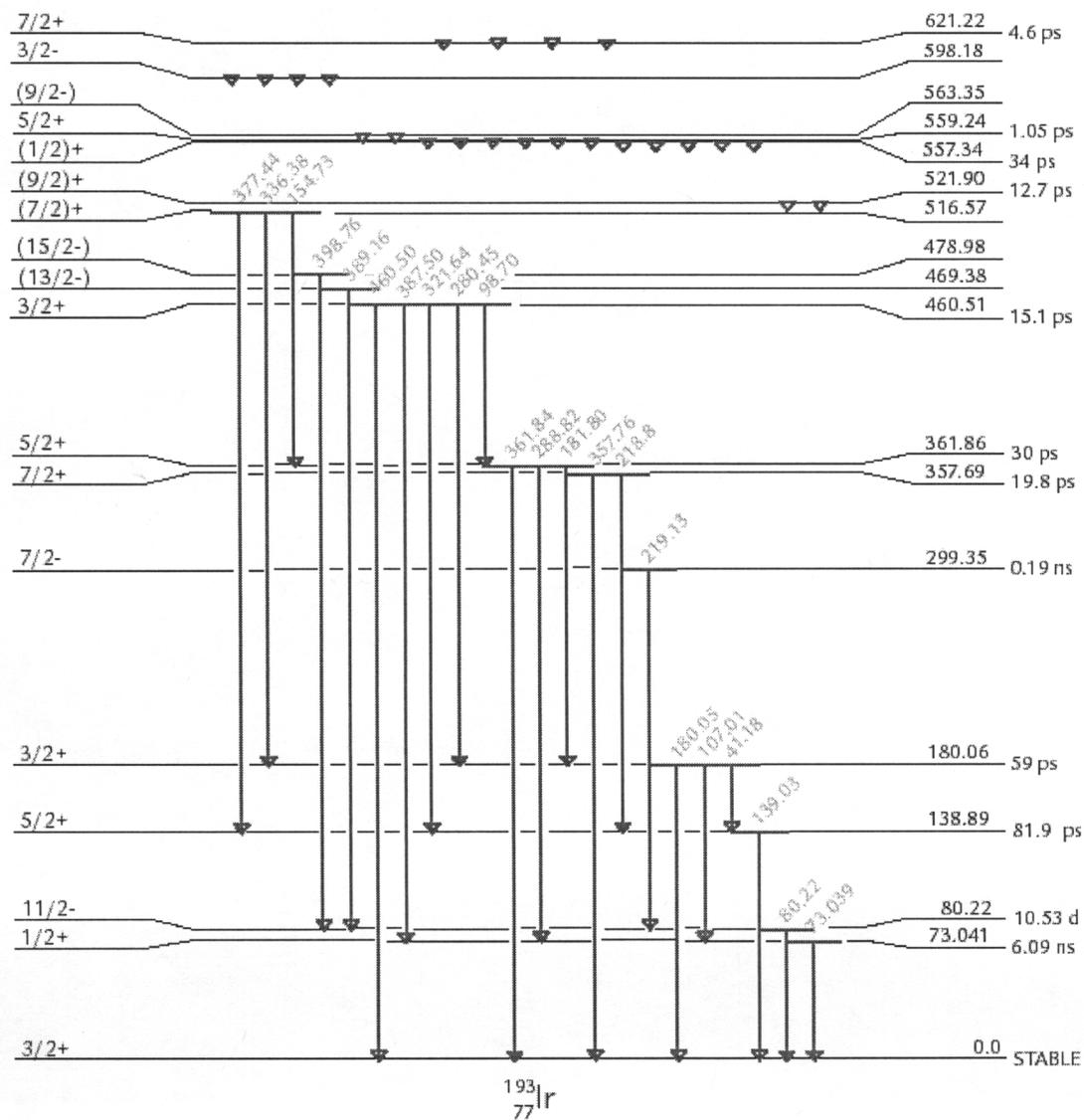


Fig. 1. Level scheme of ^{193}Ir with γ -ray cascades from decays of these excited states. The isomeric state considered in these studies has an excitation energy of 80.22 keV and a half-life of 10.53 days. [Level schemes obtained from Isotope Explorer 2.23 by S.Y. Chu, H. Nordberg, R.B. Firestone, and L.B. Ekstrom; <http://ie.lbl.gov/isoexpl/isoexpl.htm>].

The $^{193\text{m}}\text{Ir}$ state is populated (i.e., excited directly and “fed” by γ -ray cascades) by the $^{193}\text{Ir}(n,n')^{193\text{m}}\text{Ir}$ reaction (i.e., inelastic scattering in which the incident neutron loses energy in exciting the ^{193}Ir nucleus). Only one set of measurements of the population of this state existed, in part, because of the difficulty in obtaining sufficiently intense

monoenergetic neutron sources at energies from 6 to 13 MeV. The previous data and an evaluation (i.e., GNASH 98) are shown in Fig. 2. The recent γ -ray-cascade data obtained on GEANIE by the LANL/LLNL team and supplemented by model calculations are also shown in Fig. 2. The LANL/LLNL results are in sharp disagreement with the GNASH 98 cross-section evaluation over most of the energy range (4 to 8 MeV) where this cross section is large.

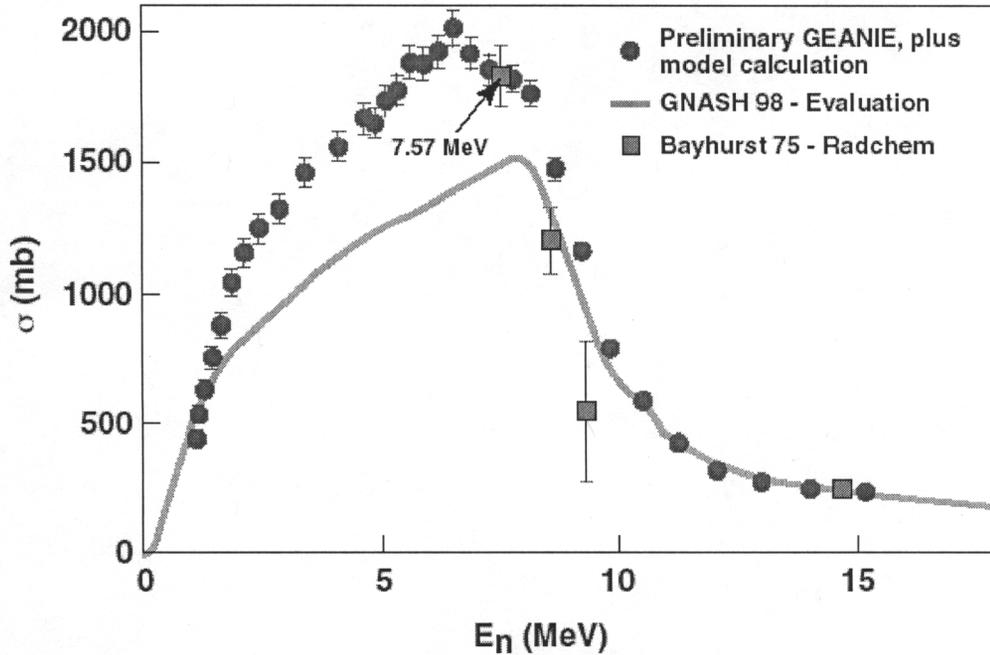


Fig. 2. Activation cross section for the $^{193}\text{Ir}(n,n')^{193\text{m}}\text{Ir}$ reaction. The GNASH 98 evaluated cross section is shown as a green curve. The experimental data from Bayhurst *et al.* are shown as solid squares. The newly obtained GEANIE results, supplemented by model calculations, are shown as solid circles.

The previous experimental data point at 7.57 MeV is in agreement with the LANL/LLNL results but far above the cross-section evaluation at this energy. The two other similar experimental data at higher energies are, however, more in agreement with the evaluation. A new evaluation will be performed using the final GEANIE results. Results for (n,2n) and other reactions on ^{193}Ir will be obtained as well.

Future measurements on ^{191}Ir similar to those done on ^{193}Ir will be carried out by the LANL/LLNL team. Studies of excited states in these two iridium isotopes are also of

interest from a nuclear structure point of view to obtain a better understanding of the shapes and excitations of these nuclei. And, as seen from the results above, better nuclear reaction modeling of the populations of the metastable states is needed.

Europium Isotopes

Natural europium consists of ^{151}Eu (47.3%) and ^{153}Eu (52.2%) — both of which can serve as radiochemical detectors. With a threshold of 7.98 MeV, the $^{151}\text{Eu}(n,2n)^{150}\text{Eu}$ reaction can produce ^{150}Eu in an isomeric state (called $^{150\text{m}}\text{Eu}$) at an excitation energy of $E_x = 42.1$ keV and with a half-life of 12.8 hours, which is compatible with commonly used radiochemical techniques (see Fig. 3).

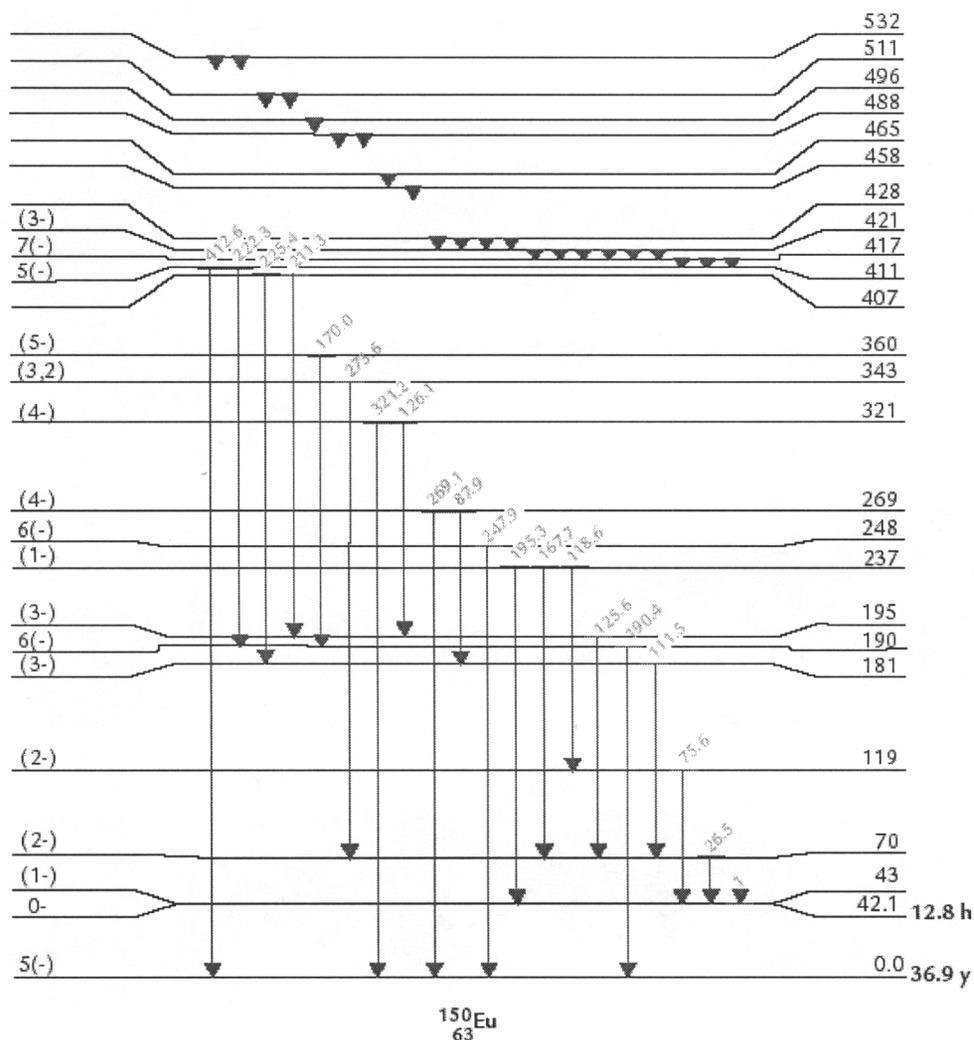


Fig. 3. Level scheme of ^{150}Eu with γ -ray cascades from decays of these excited states. The isomeric state considered in these studies has an excitation energy of 42.1 keV and a half-life of 12.8 hours. [Level schemes obtained from Isotope Explorer 2.23 by S.Y. Chu, H. Nordberg, R.B. Firestone, and L.B. Ekstrom; <http://ie.lbl.gov/isoexpl/isoexpl.htm>].

There are, however, uncertainties in the activation cross section for the production of this isomer (see Fig. 4). The $^{151}\text{Eu}(n,2n)^{150}\text{Eu}$ cross section has been measured by directly counting the two neutrons emitted in the reaction. This measurement, however, can only give the sum of the cross sections for the reactions ending with the ^{150}Eu ground state (called ^{150g}Eu , where “g” stands for “ground state”) and ^{150m}Eu because these two states are separated by an energy that is too small to be detected in the measurement.

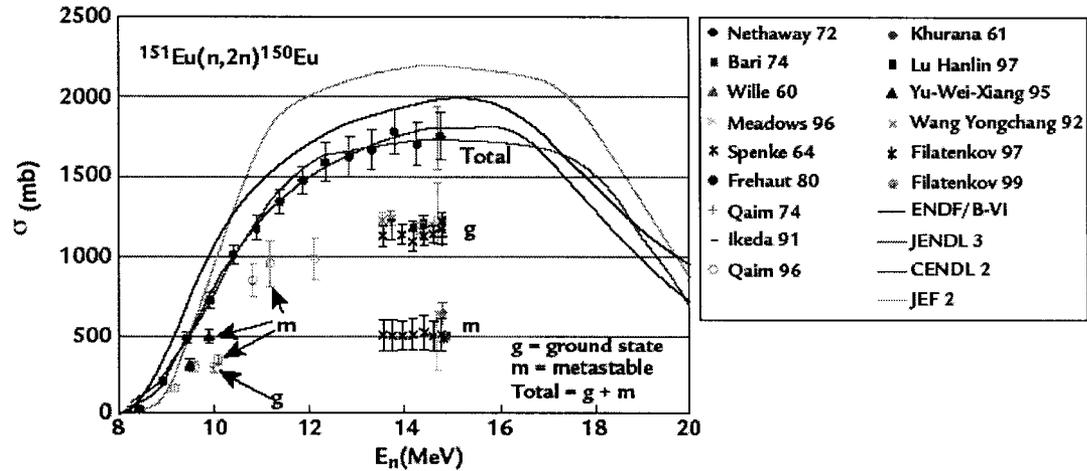


Fig. 4. Plots of $^{151}\text{Eu}(n,2n)$ cross sections for neutron energies between 8 and 20 MeV. Experimental data points are given for the $^{151}\text{Eu}(n,2n)^{150g}\text{Eu}$ cross section (g) and for the $^{151}\text{Eu}(n,2n)^{150m}\text{Eu}$ cross section (m). Data points and evaluated data (solid lines) are also given for the sum of these two cross sections (total = g + m). [Data retrieved from the CSISRS database, $^{151}\text{Eu}(n,2n)$ EXFOR files, with more references therein, available from the National Nuclear Data Center at <http://www.nndc.bnl.gov/>].

Cross sections for the $^{151}\text{Eu}(n,2n)$ reactions that lead separately to the ground and isomeric states have been measured by radiochemical methods. The sum of the $^{151}\text{Eu}(n,2n)^{150g}\text{Eu}$ and the $^{151}\text{Eu}(n,2n)^{150m}\text{Eu}$ cross sections, as obtained from the experiments and from several evaluations, is also shown in Fig. 4. The energy of interest is 14 MeV — the energy of fusion neutrons — and below to account for the degradation of neutron energy during the detonation of the device (down to the threshold energy of 7.98 MeV). There are uncertainties of $\pm 25\%$ and of $\pm 10\%$ in the $^{151}\text{Eu}(n,2n)^{150m}\text{Eu}$ and $^{151}\text{Eu}(n,2n)$ cross sections, respectively. Reducing these errors will improve the sensitivity of the diagnostics.

GEANT4 is well suited to discern the complex γ -ray cascades in ^{150}Eu — an odd-N/odd-Z nucleus. Of particular concern is the doublet of excited states at 42.1 keV (^{150m}Eu) and 43 keV (see Fig. 3) whose γ -ray feeding must be resolved to correctly deduce the

$^{151}\text{Eu}(n,2n)^{150g}\text{Eu}$ and the $^{151}\text{Eu}(n,2n)^{150m}\text{Eu}$ cross sections. GEANIE's high resolution makes it possible to detect and separate the specific γ -ray cascades that feed the ^{150m}Eu and ^{150g}Eu levels. Moreover, knowing the formation by neutron-inelastic scattering of ^{151}Eu in its ground state and its main isomeric state at 196.245 keV, with a half-life of 58.9 μs , will allow a full computer simulation of the production of the isomeric state of ^{150}Eu . Such a calculation will include the effect of the ^{151}Eu isomeric state for the first time.

The LLNL/LANL team has collected data on the γ -ray spectra emitted in the reactions described above and are currently in the process of analyzing these data. Preliminary results are shown in Fig. 5.

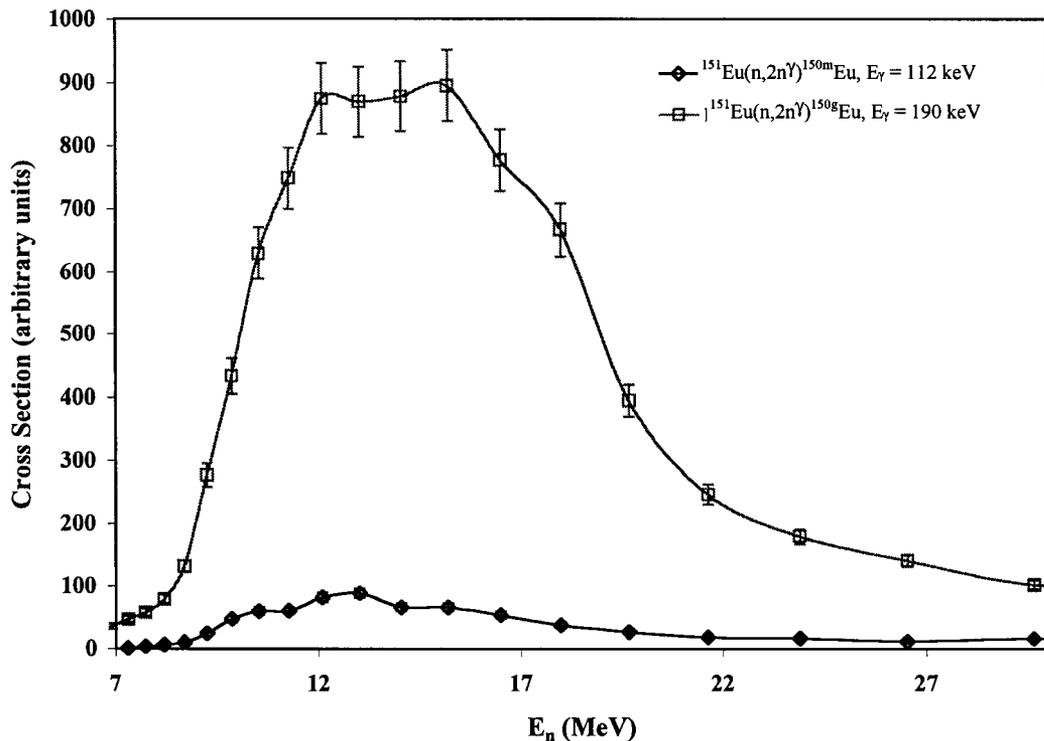


Fig. 5. Preliminary excitation functions for two of the stronger transitions observed in the $^{151}\text{Eu}(n,2n)^{150}\text{Eu}$ measurement. The 190-keV transition directly populates the ground state (squares); the 111.5-keV transition populates the long-lived isomeric state through the 70-keV excited state (diamonds).

Conclusion

Combining γ -ray-cascade measurements obtained on GEANIE with nuclear-reaction model calculations allows scientists to determine activation cross sections for $^{193}\text{Ir}(n,n')^{193m}\text{Ir}$ and $^{151}\text{Eu}(n,2n)^{150m}\text{Eu}$ reactions and other cross sections useful for radiochemical diagnostics. Once these activation cross sections are accurately known, iridium and europium isotopes could be used as diagnostics to help scientists better understand the detonation of nuclear devices, as well as in other applications for which radiochemical-fluence measurements are needed. These types of data have been difficult to obtain with "standard" techniques in many cases. An added bonus is that, with

GEANIE at WNR, the data extend to much higher incident-neutron energies (up to ~ 200 MeV) such as those found in systems for transmutation of nuclear waste or other high-energy systems that use neutrons.

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