Determination of partial (n,xnγ) cross-sections in ^{193}Ir

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Abstract

The $^{193}Ir(n,n')^{193m}Ir$ cross section for the production of the 80-keV isomer in ^{193}Ir is evaluated using a combination of experimental data and nuclear reaction modeling, from threshold to about 20 MeV. Four discrete γ lines feeding the isomer were recently observed with the GEANIE γ-ray detector at LANSCE. Theoretical calculations of the nuclear reaction mechanisms in play are then carried out to evaluate the contributions not accounted for in the experimental setup (direct population; fraction of γ-lines not observed in the experiment; etc). Experiment and modeling are then combined to provide a total cross section for the production of the Iridium isomer. We finally compare our result with activation measurement data available for a few energy points.

1 Introduction

Radiochemical detectors are commonly used to probe various energy components of a neutron fluence in a given setting. Iridium-191,193 isotopes constitute a particularly interesting set of detectors: the few-MeV region can be probed by the (n,n') production of the 80-keV (11/2-, 10.5 days) isomer in ^{193}Ir, while the higher-energy part of the spectrum can be assessed through higher-order (n,xn) reactions on both ^{191,193}Ir isotopes. This can be compared with the plutonium $(n,2n)$ cross section whose excitation function peaks near 11 MeV, and which has been recently successfully evaluated [1], using an approach similar to the one described in the present work.

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Only one experimental measurement of the $^{193}Ir(n,n')^{193m}Ir$ isomer population cross section, by Bayhurst et al. [2], was available prior to the present work. These are foil activation measurements obtained for four energies only: 7.57, 8.59, 9.34, and 14.7 MeV. In the present experiment, the GEANIE γ-ray detector was used to obtain the sum of four, well resolved, individual γ-rays feeding the isomer, and this for a wide range of incident energies from threshold to > 20 MeV.

Nuclear reaction modeling can be used to calculate the same quantity, i.e., the sum of the 4 individual γ-rays, along with the total isomer population cross section. The ratio of these two quantities is then used to correct the experimental data by accounting for the missing contributions:

$$\sigma(n,n') = \sigma^{GEANIE}(n,n'\Sigma 4\gamma) \times \frac{\sigma^{theory}(n,n')}{\sigma^{theory}(n,n'\Sigma 4\gamma)}. \qquad (1)$$

In the following sections, the GEANIE experiment is first described, followed by an explanation of the key components entering into the theoretical modeling of this isomer cross section. Finally, preliminary results are presented and discussed in view of other experimental data available. In particular, the role played by integral data coming from critical assemblies experiments is introduced.

2 GEANIE Experiment

The experiment was performed using the GEANIE escape-suppressed germanium detector array consisting of 15 coaxial HPGe detectors and 11 planar ($LEPS$) HPGe detectors [3]. GEANIE is sited on a well-collimated, 20.34 m-long neutron flight path at the WNR facility at the Los Alamos Neutron Science Center [4]. Incident neutron energy is determined by the time-of-flight technique. The useful incident neutron energy range covered from 150 keV to over 200 MeV.

The sample consisted of 2.0 grams of Ir metal powder, enriched 99.59% in ^{193}Ir purchased from the ORNL Isotope Sales Office. The Ir powder was tightly encapsulated in a thin-walled polystyrene capsule with an inner diameter of 1.265 cm and a length of 0.34 cm. The Ir sample was smaller than the neutron beam spot and centered in the beam.

For normalization purposes, two thin disks of 99.5% pure 0.005 cm thick Fe foils of 1.27 cm diameter were placed at one end of the Ir capsule during part of the experiment. The Ir data were normalized to the Fe 847-keV gamma ray cross section at 14 MeV. Corrections were made for attenuation of the gamma-rays from the Ir and from the Fe using the Monte Carlo code MCNP [5] with a detailed model of the sample and detectors.

The neutron fluence was measured with a fission ionization chamber having ^{238}U and

^{235}U fission foils. [6] The data were binned into suitable neutron energy bins. The gamma ray yields were obtained using the peak-fitting code GF3. The absolute partial γ-ray cross sections were obtained using the relation:

$$\sigma_\gamma(E_n) = \frac{I_\gamma \quad DeadT_\gamma \quad (1+\alpha_\gamma)}{\Phi(E_n) \quad DeadT_\Phi \quad \epsilon_\gamma \quad t} \tag{2}$$

where σ_γ is in mbarn;

I_γ is the yield of a γ ray in counts/MeV;

Φ is the incident neutron fluence in neutrons/MeV;

ϵ_γ is the detection efficiency of the array for the particular γ-ray energy corrected for attenuation within the target using MCNP [5];

α_γ is the conversion coefficient for the particular γ ray;

t is the target thickness in atoms/mb;

$DeadT_\gamma$ and $DeadT_\Phi$ are the dead time corrections for the detection of γ rays from the array and neutrons in the fission chamber, respectively.

A total of 160 gamma rays from reactions ranging from (n, n') to $(n, 11n)$ were observed. About 60 of the gamma rays are from inelastic scattering. Of these 60 gamma rays, 18 feed the isomer at 80 keV, and the remainder decay to the ground state.

3 Theoretical Modeling

Our nuclear reaction model calculations were carried out with the GNASH code, described at some length in Ref. [7]. The GNASH code implements the statistical Hauser-Feshbach theory of nuclear reactions, which conserve angular momentum and parity. The population cross section of the 11/2- isomer in ^{193}Ir can indeed only be modeled accurately if careful attention is given to angular momentum transfer processes. The GNASH code follows in detail the formation of a compound nucleus, followed by its decay by particle and radiative emissions. To fully account for the physics at hand, non-statistical effects have to be added to this picture of a fully equilibrated compound nucleus. In particular, preequilibrium emission of high-energy particles and γ-rays are included through the quantum mechanical Feshbach-Kerman-Koonin (FKK) theory [8], and eventually using the HMS model by Blann and Chadwick [9, 10].

The shapes of the (n, n') and $(n, 2n)$ excitation functions above about 10 MeV depends significantly on these precompound emissions. If a significant number of high-energy neutrons is emitted, then the probability of a secondary neutron emission is much reduced.

In short, preequilibrium emissions will tend to increase the tail of the (n, n') cross section, at the expense of the $(n, 2n)$ excitation function.

At low-incident energies, the Bohr assumption of totally uncorrelated ingoing and outgoing channels fails (very small number of open channels), and so-called width fluctuation correction factors have to be included in the reaction modeling. The traditional Hauser-Feshbach formula for the cross section σ_{ab}, then reads

$$\sigma_{ab} \propto \frac{T_a T_b}{\sum_c T_c} \times W_{ab}. \tag{3}$$

The symbols a and b are shortcuts for describing ingoing and outgoing channels, with all their associated quantum numbers. The transmission coefficients $\{T_c\}$ are the result of optical model calculations. Finally, the factors W_{ab} represent the width fluctuation correction factors. In the present calculations, the Moldauer expression [11] was used to infer these correction factors as

$$W_{ab} = \left(1 + \delta_{ab}\frac{2}{\nu_a}\right) \int_0^{+\infty} dx \prod_c \left(1 + \frac{2T_c}{\nu_c \sum_i T_i} x\right)^{-\delta_{ac}-\delta_{bc}-\nu_c/2}, \tag{4}$$

where the number of degrees of freedom ν_i read

$$\nu_i = 1.78 + \left(T_i^{1.212} - 0.78\right) exp\left(-0.228 \sum_c T_c\right). \tag{5}$$

Recently, Hilaire et al. [12] showed that Moldauer's approximate formula reproduces quite well the exact result obtained by Verbaarschot [13]. Note however that this conclusion has to be softened in the limit of strong absorption [14].

Such width fluctuation corrections tend to increase the elastic cross section at the expense of the other competing channels, in particular the capture cross section. Until recently, these calculations were performed using the COMNUC code, by passing its output to the GNASH code. We have recently developed a new code, called McGNASH, which is intended to replace the GNASH code in the near future. Among the improvements over GNASH is the full implementation of the width fluctuation corrections within the Hauser-Feshbach code. Calculations with GNASH+COMNUC and McGNASH have been performed with very good agreement between them.

Of course, these reaction cross section calculations depend very much on the various models (and their input values!) used in the decay process computation: nuclear level densities, optical model transmission coefficients, and γ-ray strength functions (particularly at low-incident energies). In this regard, available experimental data are crucial in constraining the allowed input parameters-space.

A key component in the evaluation of (n, n') and (n, xn) reaction cross sections is a reliable optical model potential, which provides the angular momentum and spin dependent transmission coefficients entering the Hauser-Feshbach decay equations. The optical model calculations were performed with the coupled-channels code ECIS [15]. In addition to providing these spin-dependent probability decay rates, ECIS also determines cross sections of the direct reactions exciting low-lying collective states. The optical potential chosen here is based on the work of P.G. Young [16] for neighboring *Re* isotopes and adapted for isospin differences to *Ir* isotopes. These coupled-channel ECIS calculations are described at some length in Ref. [17].

Nuclear level densities are also a critical ingredient in such Hauser-Feshbach calculations. The Ignatyuk et al. [18] prescription is used here, which includes a washing-out of shell effects in the level density with increasing excitation energy. The energy-dependent level density parameter is

$$a(U) = \alpha \left(1 + f(U)\delta W/U\right), \tag{6}$$

where α is the asymptotic (high-energy) parameter value. The term δW represents the shell corrections, and is defined as

$$\delta W = M_{exp}(Z, A) - M_{ld}(Z, A, \beta), \tag{7}$$

where $M_{exp}(Z, A)$ is the experimental mass and $M_{ld}(Z, A, \beta)$ is the liquid drop mass at quadrupole deformation β. The damping of shell effects with increasing energy is handled through $f(U) = 1 - exp(-\gamma U)$, with γ=0.05 MeV^{-1}. Finally, the asymptotic value α is obtained through s-wave resonance data fitting [19].

In addition to the particle decay probability coefficients obtained with the ECIS coupled-channels calculations, one needs to have information on γ decay probabilities. In the present calculations, γ-ray emission is treated within the Generalized Lorentzian strength function formalism of Kopecky and Uhl [20]. Only E1, M1, and E2 transitions are considered. When available, experimental data on $2\pi \langle\Gamma_0\rangle \,/\, \langle D_0\rangle$ can be used to renormalize the various strength functions. For further details, see Ref. [17].

4 Results and Discussion

GEANIE data and GNASH calculations for the four observed γ-rays are compared in Fig. 1. First, the shapes of these four experimental excitation functions appear quite well reproduced by theoretical calculations. For three of the four γ-lines, the calculations are even in fair quantitative agreement with the experimental points. This tends to demonstrate that angular momentum transfers in the decay process are well handled by the calculations. However, large discrepancies occur for the highest-spin (15/2-), E_γ = 398.8 keV line. These differences still need to be resolved, but may be due to an incomplete knowledge of the nuclear levels involved in the γ decay.

Applying Eq. 1 only to the three discrete lines well reproduced theoretically, we can now obtain the total isomer production cross section. The result is depicted in Fig. 2. The

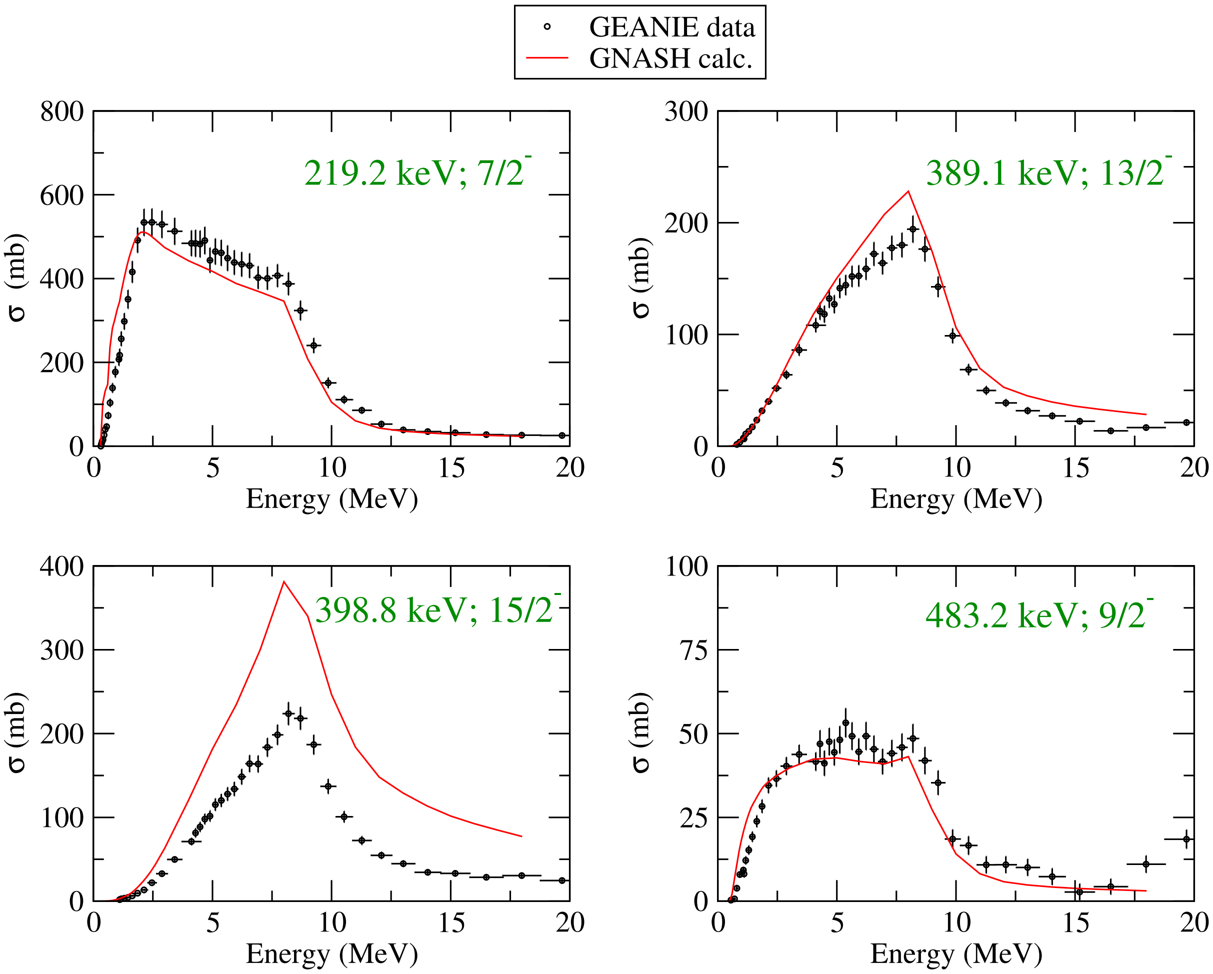

Figure 1: GEANIE data vs. model calculations for the four observed γ-rays feeding the 80 keV isomer in ^{193}Ir.

GEANIE data points, corresponding to the sum of the three discrete lines, are represented by circles, while the GEANIE+GNASH results appear as squares. The latter can then be compared to the activation measurements by Bayhurst, and to the purely theoretical GNASH calculations for the total (n,n') cross section.

First of all, the GNASH calculation agrees fairly well with the GEANIE+GNASH result. This is not surprising since the three individual measured excitation functions were already well accounted for by GNASH calculations (see Fig. 1). Obviously, the GEANIE+GNASH points would deviate from the GNASH result if the fourth γ-ray line (E_γ=398.8 keV) would be included in this final analysis.

A second interesting point is that the GEANIE+GNASH points are in fair agreement with the Bayhurst activation measurement. In particular, the 14 MeV point is quite well reproduced, although the present results do not agree with the 7.57 MeV point by Bayhurst.

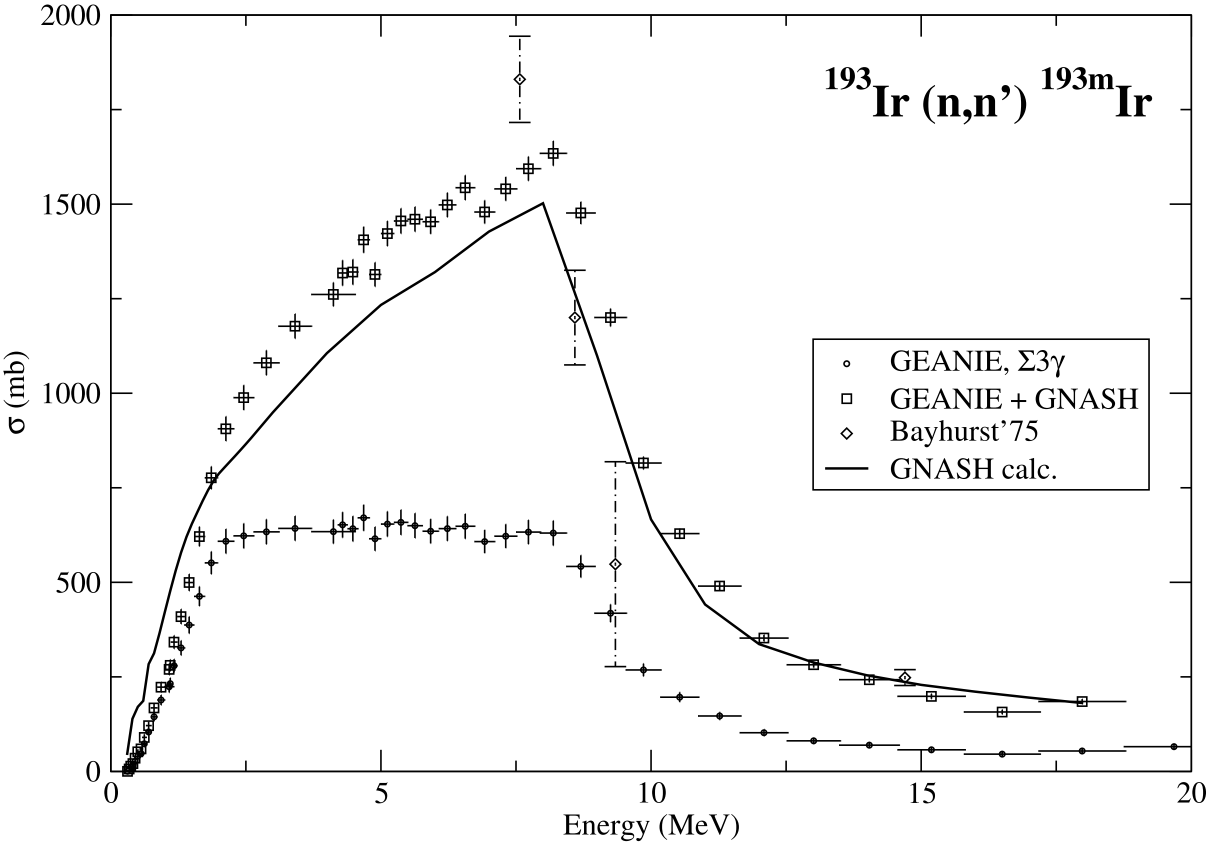

Figure 2: Experimental data corrected by model calculations to take into account missing contributions. Black points represent the GEANIE data for the sum of three observed γ-lines; the red points correspond to the sum discrete lines corrected by model calculations using Eq. 1. These points can be compared with activation measurement data by Bayhurst et al. [2].

A closer look at this excitation function however reveals some disturbing discrepancies. In particular, the calculated low-energy part of the spectrum, from threshold (299.2 keV) to about 2 MeV, largely overestimates the GEANIE + GNASH points. In this region, one would expect theoretical predictions to be more accurate since less energetic neutrons are exciting low-lying states only, and therefore complications arising because of complex level density distributions are eliminated.

A key tool to help us investigate this low energy region in more details may come from integral experiments such as the available critical assemblies information. In such assemblies, a particular neutron spectrum is present, and various cross sections of interest are measured in an integral fashion. As an example, the $^{193}Ir(n,n')^{193m}Ir/^{191}Ir(n,\gamma)^{192}Ir$ ratio was measured relative to $^{238}U(n,f)/^{235}U(n,f)$, the latter roughly corresponding to a measure of the hardness of the neutron spectrum. The data coming from such an experiment is called integral because the cross sections are not directly measured, but instead folded with the neutron spectrum at hand. Such data can be very useful in guiding our evaluation. In particular, the low-energy rise of the 80 keV-isomer cross section can be assessed through these critical assemblies data. This work is currently underway.

5 Conclusion

We have studied, both theoretically and experimentally, the (n, n') production cross section of the 80-keV (10.5 days) isomer in ^{193}Ir. Peaking in the few-MeV region, this excitation function is of great interest in radiochemical analyses to probe fission neutrons fluences.

Prior to this work, only one measurement by Bayhurst et al. [2] using a foil activation technique had been performed, and in particular nothing was known below $\sim$7 MeV incident neutron energy.

By combining the recent GEANIE experimental data and GNASH nuclear reaction modeling calculations to take into account experimentally missing contributions, we obtained a new evaluation for this isomer population cross section over the entire energy range from threshold ($\sim$300 keV) to $>$ 20 MeV.

Although some discrepancies between experiment and calculations have not been resolved yet, our preliminary results are very encouraging and should finally lead to a tremendous improvement over past evaluations. Finally, some additional experimental integral data should help us in constraining even more our final evaluated cross section.

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