Exploring the origin of the rarest stable isotopes naturally occurring on Earth with real photon beams

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Abstract. This lecture brings into focus research work that aims to advance fundamental knowledge on a forefront topic in nuclear astrophysics – the nucleosynthesis beyond Fe of the rarest stable isotopes naturally occurring on Earth (the origin of *p*-nuclei). The astrophysical phenomenon responsible for this synthesis is termed the *p*-process. Though modelling the *p*-process nucleosynthesis is a daunting task, significant progress can be made by performing key experimental studies, which can constrain Hauser-Feshbach statistical nuclear models used to calculate the many unknown astrophysically relevant stellar reaction rates. Photodisintegration stellar reactions play a major role in the *p*-process; they are especially highly sensitive to the low-energy tail of the nuclear photon strength function (PSF). Recent experimental efforts to constrain pSFs for the *p*-process nucleosynthesis calculations using real photons at the HIγS/TUNL facility and at the Madison Accelerator Laboratory (MAL), an unconventional bremsstrahlung facility that features a repurposed medical electron linear accelerator, will be discussed in this lecture.

1 Introduction

The astrophysical *p*-process is responsible for the nucleosynthesis beyond iron of about 35 proton-rich stable nuclei, the so-called *p*-nuclei [1]. As they are located on the neutron deficient side of the valley of β-stability, they cannot be synthesized by radiative neutron capture reactions; hence, they are shielded from the s- or r-process. In the solar system, the *p*-nuclei are one or two orders of magnitude less abundant than the *s*- or *r*-nuclei [2]. The astrophysical details of the *p*-process are still under discussion. So far, it has been impossible to reproduce solar abundances of all *p*-nuclei in a single process, but it is generally accepted that the main mechanism (also known as the γ-process) to produce *p*-nuclei involves photodisintegration reactions on pre-existing s/r-seed nuclei and β+ decays and that it occurs mainly in explosive O/Ne burning during supernova type II explosions at temperatures in the range of 2 – 3.5 GK.

Modelling the *p*-process nucleosynthesis is a daunting task, requiring an extended network of about 20000 nuclear reactions linking about 2000 nuclei [1]. Obviously, it is not

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feasible to measure all the required cross sections, thus we will have to rely on theoretical calculations of the many unknown stellar reaction rates of interest.

The laboratory ground state contribution to the stellar rate for photodisintegration reactions of interest for the *p*-process is only a few tenth per mille [3, 4]. Therefore, photodisintegration cross section measurements can only be used to derive information on certain nuclear properties (such as photon strength functions (PSFs), nuclear level densities, nucleon-nucleus optical potentials) required for the calculation of the stellar rates and, thus, to test and validate the theory (statistical Hauser-Feshbach nuclear models).

In the following we discussed our recent efforts to constrain PSF models via two distinct experimental methods by studying in the laboratory photonuclear reactions using real photon beams produced at the High Intensity γ-ray Source (HIγS) facility [5] at Duke University, Durham, North Carolina, USA and at the Madison Accelerator Laboratory (MAL) [6] at James Madison University, Harrisonburg, Virginia, USA.

2 Photoneutron reaction cross section measurements on ^{94}Mo and ^{90}Zr relevant for the *p*-process nucleosynthesis

The most abundant *p*-nuclei, ^{92,94}Mo and ^{96,98}Ru, are notoriously underproduced in the currently favoured scenarios for the *p*-process [1], making their nucleosynthesis a longstanding mystery in nuclear astrophysics. Recent studies [7, 8] of type Ia supernovae with two-dimensional models (deflagration and delayed detonation) demonstrated that both light and heavy *p*-nuclei, including the much-debated ^{92}Mo and ^{96,98}Ru, are produced with similar enhancement factors relative to solar abundances, provided an *s*-process enrichment of the progenitor is assumed. However, ^{94}Mo remained underproduced in these studies. Remarkably, ^{90}Zr, known as a genuine *s*-process nuclide, was significantly produced in these studies. In light of these intriguing findings, we were motivated to investigate the photoneutron reactions on ^{94}Mo and ^{90}Zr using real photon beams produced at the HIγS facility. We measured excitations functions from slightly above the respective neutron separation energies up to 13.5 MeV with the goal of extracting experimentally the PSFs to be used for calculation of photonuclear stellar reaction rates for the *p*-process nucleosynthesis.

Experimental details and our final findings regarding how sensitive the ^{94}Mo(γ,n) and ^{90}Zr(γ,n) stellar reaction rates are to the corresponding measured cross sections can be found in our recently paper [9]. Here, we highlight an important experimental conclusion that we can draw from our measurements with respect to the neutron detection efficiency.

In our experiment the neutrons were detected using an assembly of 18 tubular proportional counters filled with ^{3}He gas at a pressure of ~6 atm and embedded in a cylindrical polyethylene body that served as a neuron moderator. For the energies studied in our work, the ^{94}Mo(γ,n) and ^{90}Zr(γ,n) reaction only proceed directly to the ground state of their respective residual nuclei at low beam energies. At higher energies, population of the ground state in the residual nuclei proceeds predominantly via the population of excited states which then γ decay to the ground state. Because the information on the neutron energy is lost by the thermalization of the neutrons in the moderator, determining the neutron detection efficiency for such a detector was a complex problem to tackle. The neutron detection efficiency depends on the neutron energy. Neutron energies in our work span a broad range from 20 keV up to ~4 MeV that correspond to detection efficiencies as high as ~55% and a slow as ~25%, respectively. These efficiencies were simulated in GEANT4 assuming isotopically distributed neutrons.

In the case of the ^{94}Mo(γ,n) reaction, Fig. 1 shows that there is a good agreement between our cross-section results and the previous results by Utsunomiya *et al.* [10] and by Beil *et al.*

[11] for photon beams below 10.8 MeV. However, above that beam energy, when neutrons are emitted predominantly from excited states in ^{93}Mo, there is a significant discrepancy between our results and previous results as high as ~30%. In Fig. 1 we also show that our results become in reasonable agreement with the previous work if the neutron detection efficiency is calculated assuming that all detected neutrons are ground state neutrons. The conclusion that we can draw from here is that measured neutrons that are correlated with excited states in the residual nucleus must be appropriately accounted for when determining the detection efficiency needed to extract the laboratory photoneutron cross sections – *if only ground state neutrons are considered, the detector efficiency will be underestimated and consequently the photoneutron reaction cross section will be overestimated.*

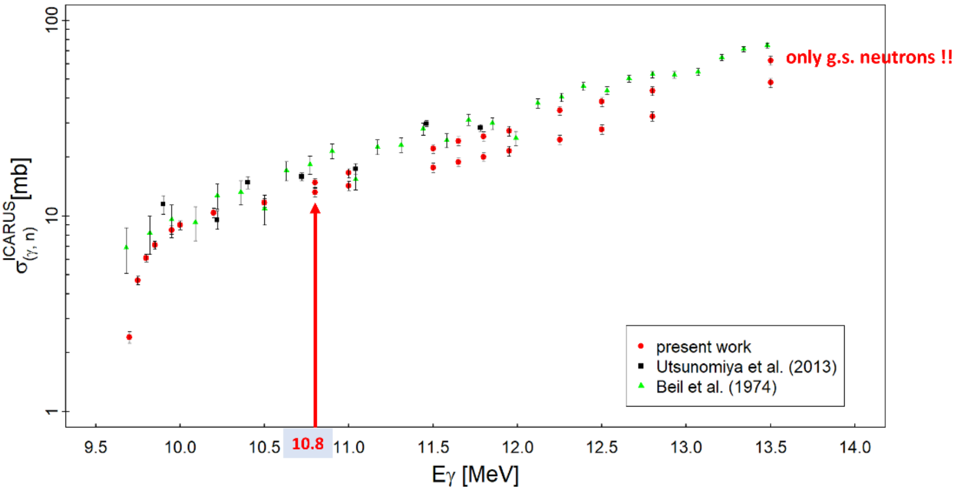

Fig. 1. Excitation function for ^{94}Mo(γ,n) of our work compared with previous work. See text for details and Ref. [9].

To address this discrepancy between our work [9] and previous work [10, 11], new measurements using *flat-efficiency neutron detectors* [12] will be the best call of action.

3 Exploring the origin of the *p*-nuclei via photon-induced activation studies at the Madison Accelerator Laboratory

The focus of this research project is to determine experimentally the ground state reaction rates for eight photoneutron reactions to be investigated via photon-induced activation at the Madison Accelerator Laboratory (MAL), using bremsstrahlung photons from an electron linear accelerator by applying the so-called *superposition method* [13]. This experimental technique has been established and well tested allowing the determination of the ground-state photodisintegration reaction rates directly from bremsstrahlung induced activation experiments.The eight photoneutron reactions of interest are: **^{64}Zn(γ,n)^{63}Zn, ^{70}Ge(γ,n)^{69}Ge, ^{74}Se(γ,n)^{73}Se, ^{78,80}Kr(γ,n)^{77},^{79}Kr, ^{84,86}Sr(γ,n)^{83,85}Sr and ^{90}Zr(γ,n)^{89}Zr**.

Photoactivation studies using bremsstrahlung as real photon sources have been widely performed at conventional linear accelerators around the world [13], most notably at the superconducting Darmstadt linear electron accelerator S-DALINAC [14] and at the superconducting electron accelerator ELBE of the Forschungszentrum Rossendorf [15]. However, using an “off-the-shelf” medical linear accelerator, originally designed for clinical operation and repurposing it for nuclear physics experiments, is a novel idea.

The general experimental setup of photoactivation experiments, obviously limited to reactions where an unstable nucleus is produced, is fairly simple. A target sample (made of stable nuclei) is irradiated with bremsstrahlung photons. After the activation, the produced γ-ray activity is counted offline with a high-purity germanium (HPGe) detector, well-shielded against (natural) background. The photoactivation technique, has been chosen for the experiments using bremsstrahlung photons because a direct detection of the emitted particle is difficult in the huge bremsstrahlung background. Additionally, the excellent energy resolution of γ-ray detectors allows a clear detection of the individual reactions even in a chemically mixed target with natural isotopic composition.

The reaction rate $\lambda_{(\gamma,j)}$ for a photoreaction induced on a ground-state nucleus, leading to the emission of particle *j*, is given by the expression:

$$\lambda_{(\gamma,j)} = \int_0^\infty c n_\gamma(E) \sigma_{(\gamma,j)}(E) dE \tag{1}$$

where *c* is the speed of light, $\sigma_{(\gamma,j)}$ the cross section and $n_\gamma(E)$ the number of photons per unit volume and energy *E*. In a stellar interior at a temperature *T*, $n_\gamma(E)$ is very close to the black-body or Planck distribution:

$$n_\gamma^{Planck}(E,T) = \frac{1}{\pi^2} \frac{1}{(\hbar c)^3} \frac{E^2}{\exp(E/kT) - 1} \tag{2}$$

The aforementioned *superposition method* approximates the high energy tail of the Planck distribution of Eq. (2) by a superposition of several bremsstrahlung spectra with different endpoint energies. This is illustrated in Fig. 2 for *T*= 2.5 GK, a typical *p*-process temperature, using six bremsstrahlung spectra with endpoint energies E_{max} from 8 to 10 MeV. Depending on the temperature *T* the bremsstrahlung spectra, calculated and correctly described by Monte Carlo simulations using the code GEANT4 have to be weighted by factors $a_i(T)$. This leads to the following equation for the approximation applied in the superposition method:

$$c n_\gamma^{Planck}(E,T) \approx \sum_i a_i(T) \Phi_\gamma^{brems}(E, E_{\max,i}) \tag{3}$$

where $\Phi_\gamma^{brems}(E, E_{\max,i})$ is the photon flux in the bremsstrahlung spectrum with endpoint energy $E_{max,i}$.

Combining Eqs. (1) and (3) astrophysical *ground-state* reaction rates for (γ,n) photodisintegrations can be determined experimentally from the relation

$$\lambda_{(\gamma,n)}^{gs}(T) \approx \sum_i a_i(T) \int_{E_{thr}}^{E_{\max,i}} \Phi_\gamma^{brems}(E, E_{\max,i}) \sigma_{(\gamma,n)}(E) dE \tag{4}$$

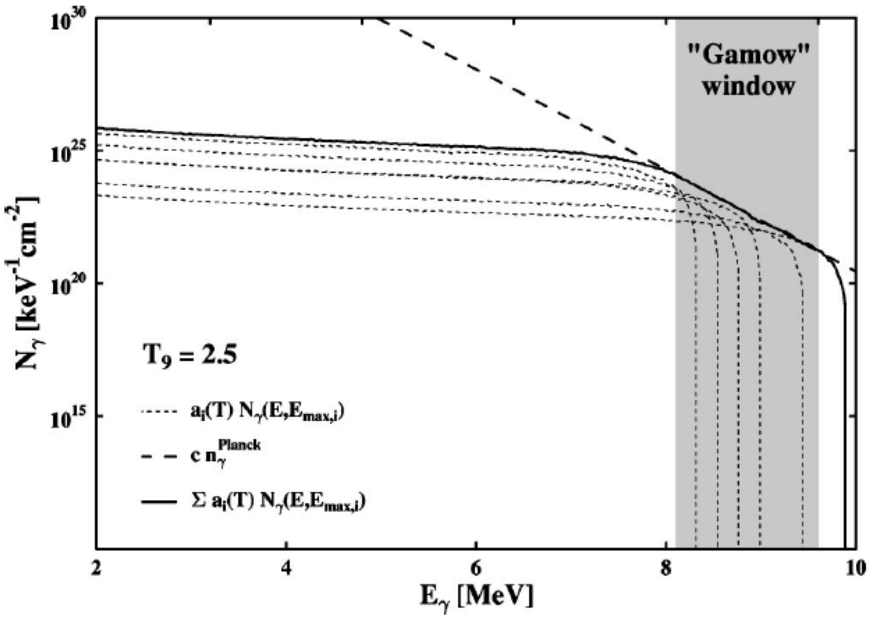

Fig. 2: Approximation of the Planck distribution by bremsstrahlung spectra. The grey shaded area corresponds to the Gamow-type energy window for (γ,n) reactions of interest for the *p*-process. See text for details.

One notes that the integral in Eq. (4) corresponding to the *i*-th bremsstrahlung irradiation of the target sample represents the energy-integrated photoneutron cross section $I_{\sigma,i}$, which is directly proportional with the reaction yield measured using the photoactivation technique. Hence Eq. (4) becomes

$$\lambda^{gs}_{(\gamma,n)}(T) \approx \sum_i a_i(T) I_{\sigma_{(\gamma,n)},i} \qquad (5)$$

The reaction yield $Y_{(\gamma,n)}$ representing the number of (γ,n)-reactions in the target during each activation run and is related to the reaction cross section:

$$Y_{(\gamma,n)} = N_t I_{\sigma_{(\gamma,n)}} \qquad (6)$$

with N_t the number of target atoms. Moreover, the reaction yield can be calculated from the number of observed γ-ray decays A_γ (determined from the offline counting with a ultra-low background HPGe) associated with the particular (γ,n) photodisintegration reaction:

$$A_\gamma = \varepsilon_\gamma I_\gamma f\left(t_{live}/t_{real}\right) Y_{(\gamma,n)} \qquad (7)$$

where ε_γ is the γ-ray detector efficiency, the I_γ absolute intensity of the observed γ decay line, t_{live}/t_{real} the dead time correction. The factor *f* accounts for the continuous decay of produced unstable nuclei during and after activation as well as during the off-line *γ*-spectroscopy measurement:

$$f = e^{-\lambda t_c}\left(1-e^{-\lambda t_m}\right)\left(1-e^{-\lambda t_{irr}}\right) \quad (8)$$

where t_c is the cooling time between the end of irradiation and the beginning of the offline measurement, t_m is the duration of the offline measurement, and t_{irr} the duration of the irradiation.

For the superposition method described earlier to determine the ground state (γ,n) reaction rates, it is necessary to measure the endpoint energy of the bremsstrahlung spectra precisely. We have been working to employ the same procedure as the procedure implemented at the bremsstrahlung ELBE facility [16] that is based on the spectroscopy of protons resulted from the photodisintegration of the deuteron – the ^{2}H(γ,p) reaction. From the pure two-body kinematics, the energy of the incident photons can be deduced directly from the measured energy of the emitted proton. The protons from the photodisintegration of the deuteron are detected by a setup of 3 silicon detectors (type ORTEC ULTRA) with an appropriate thickness so that the protons are completed stopped in the detectors.

Thus far, we have assembled a similar experimental setup as the one at ELBE [16], and currently we are in process of understanding our proton detection spectra, which seems to be dominated by electron and beam induced background.

As a byproduct of our efforts under development to measure the endpoint energy bremsstrahlung, the half-lives of five proton-rich radioisotopes that belong to a mass region relevant to the astrophysical *p*-process—^{69}Ge, ^{73}Se, ^{83}Sr, ^{85m}Sr, and ^{63}Zn—were measured with high precision via photon-induced activation at MAL. While our results are in good agreement with the respective literature adopted values, we were able in the case of ^{73}Se to improve significantly on the experimental accuracy. For details see our recent paper [17].

These results enable a more accurate determination of related ground-state photoneutron reaction rates for ^{70}Ge(γ,n), ^{74}Se(γ,n), ^{83}Sr(γ,n), ^{86}Sr(γ,n), ^{64}Zn(γ,n), which can be useful in constraining statistical nuclear models needed to calculate stellar nuclear reaction rates relevant for understanding the astrophysical *p*-process nucleosynthesis.

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