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Direct and Indirect Measurements of Neutron Induced Cross Sections at Storage Rings

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Neutron-induced cross sections of short-lived nuclei are highly relevant in many domains such as fundamental nuclear physics, astrophysics and applications in nuclear technology. In particular, these cross sections are essential for understanding the synthesis of elements via the s- and r stellar processes. However, the measurement of such cross sections with current techniques is very difficult or even impossible, because of the difficulties to produce and handle the necessary amounts of radioactive nuclei. Reaching the nuclei of interest is only possible by inverting the reaction kinematics with radioactive beams.

In this contribution we present a project for indirectly determining neutron cross sections via the surrogate-reaction method. This project is based on the measurement of transfer- or inelastic-scattering-induced decay probabilities in inverse kinematics at storage rings. The measured probabilities are then used to tune nuclear-reaction models that will provide much more accurate predictions of the desired neutron cross sections. We also discuss a very ambitious, long-term project to directly measure neutron cross sections in inverse kinematics. It consists in the combination of a radioactive beam facility, an ion storage ring and a spallation neutron source.

KEYWORDS: Neutron-induced reaction cross sections, Short-lived nuclei, Storage rings, Surrogate-reaction method, Free neutron target

1. Introduction

The synthesis of heavy elements beyond ^{56}Fe takes place primarily by neutron capture on lighter seed nuclei in the s- (slow neutron capture) and the r- (rapid neutron capture) processes. Both processes consist in consecutive radiative neutron captures and beta decays but they run in environments with very different conditions (neutron flux and temperature). Current efforts address challenges such as identifying the sites of the r-process and working out the details of the known s-process environments. For this purpose, detailed models of these processes are developed to formulate predictions for the isotopic abundances that can be compared to astrophysical observations. Neutron cross sections for radiative capture (n,γ) on unstable nuclei are the most critical input information required by these models [1]. Spontaneous and neutron-induced fission of very neutron-rich heavy nuclei also play an essential role during r-process nucleosynthesis in neutron-star mergers, as they determine the end of the r-process path and the fission products have a strong influence on the associated nuclide abundances [2]. In this case, the relevant properties are neutron-induced fission cross sections (n,f), barrier heights and fission-fragment distributions. Neutron cross sections on radioactive nuclei are also relevant for the development of advanced nuclear systems for sustainable energy production and nuclear waste management [3], as well for the optimisation of the production of diagnostic or therapeutic radionuclides for nuclear medicine and industrial quality control [4].

The measurement of neutron cross sections of short-lived nuclei is very complicated or even impossible due to the radioactivity of the samples involved. This issue can be solved by inverting the reaction kinematics with radioactive beams. The possibility to perform nuclear-reaction measurements at storage rings that was recently demonstrated at the ESR [5] opens up new, largely unexplored possibilities for the development of indirect and direct methods for the measurement of neutron cross sections in inverse kinematics. In section 2, we present a project to indirectly infer these cross sections by measuring in inverse kinematics the decay probabilities induced by surrogate or alternative reactions that lead to the nuclei formed in the desired neutron-induced reaction. The measured decay probabilities are particularly valuable observables to understand the de-excitation process of nuclei. Therefore, they can be used to precisely tune model parameters that will lead to significantly improved predictions of the desired neutron cross section. In section 3, we describe a long-term project to directly measure neutron cross sections in inverse kinematics. The proposed facility consists in the coupling of a radioactive beam facility, a storage ring and a spallation source.

2. Indirect measurements of neutron cross sections with surrogate reactions at storage rings

2.1 Surrogate reactions

The neutron cross sections of interest populate nuclei with excitation energies of about 10 MeV. At these excitation energies the excited nucleus mainly de-excites via different competing decay channels: γ-ray emission, particle emission and fission, if the nucleus is heavy enough. Neutron-induced reactions at these energies generally proceed through the formation of a compound nucleus, i.e. a nucleus in statistical equilibrium. The cross section for a neutron-induced reaction can then be written as:

$$\sigma_{n,\chi}(E_n) = \sigma^{CN}(E_n) \cdot P_{n,\chi}(E^*) \tag{1}$$

where $\sigma^{CN}(E_n)$ is the cross section for the formation of a compound nucleus (CN) after the absorption of a neutron of incident energy E_n, E^* is the excitation energy and $P_{n,\chi}$ is the probability that a compound nucleus formed after neutron absorption decays through channel χ. The idea behind the surrogate-reaction method is to infer neutron cross sections of short-lived nuclei by calculating the σ^{CN} with the optical model and replacing $P_{n,\chi}$ in eq. (1) by the decay probability induced by a surrogate reaction $P_{s,\chi}$ that leads to the same CN, but that is much easier to realise than the neutron reaction of interest. Surrogate reactions are typically transfer or inelastic-scattering reactions. The angular momentum J and parity π of the CN formed in the neutron-induced and surrogate reactions can be very different. This can lead to significant differences between $P_{n,\chi}$ and $P_{s,\chi}$, since the decay probabilities P might strongly depend on J^{π}. Therefore, there may be cases where there are important discrepancies between directly measured neutron cross sections and the cross sections derived using surrogate reactions.

A number of examples involving fission probabilities of odd-A and odd-odd fissioning nuclei not far from ^{238}U, e.g. [6, 7] show that the fission probabilities induced by surrogate reactions are rather close to the corresponding neutron-induced fission probabilities, i.e $P_{s,f} \approx P_{n,f}$. This indicates that fission probabilities are rather insensitive to J^{π}. Thus, fission probabilities induced by surrogate reactions can be used to provide neutron-induced fission cross sections of short-lived nuclei rather directly, by simply replacing $P_{n,f}$ by the measured probability $P_{s,f}$ in eq. (1). There is no clear explanation for this finding yet and further studies involving fissioning nuclei with different structural properties, like even-even nuclei and nuclei near shell closures, are needed. We would like to stress also that fission probabilities induced by nuclear transfer and inelastic-scattering reactions represent the most direct observable to infer fission barriers.

The situation is very different for γ-emission probabilities P_{γ} induced by surrogate reactions. These probabilities measure the likelihood that the excited nucleus decays only by the emission of γ rays and can be related to neutron radiative capture cross sections (n,γ). The γ-emission probabilities induced by surrogate reactions $P_{s,\gamma}$ are much higher than neutron-induced ones, i.e $P_{s,\gamma} > P_{n,\gamma}$, see e.g. [8,9,7]. These significant differences have been attributed to the larger angular momenta populated in the surrogate reaction. Still, γ-emission probabilities are very useful to fix the parameters of some of the key ingredients of the statistical model like level densities and γ-ray strength functions. Once these parameters are fixed, the model can be used to provide much more accurate predictions of the desired neutron cross section. This has recently been shown by J. Escher et al. for the ^{90}Zr(n,γ) reaction [10].

2.2 Surrogate reaction studies in inverse kinematics at storage rings

The previous surrogate-reaction studies have been performed in regular or direct kinematics, where a light charged projectile interacts with a heavy target. These studies often suffer from significant limitations. One of the main issues is the unavailability of radioactive targets and the presence of target contaminants and the target backing. In addition, in direct kinematics the residues produced after γ or particle emission are stopped in the target. Therefore, the measurement of γ- or particle-emission probabilities requires detecting the γ-rays or the particles emitted during the decay. This is rather complicated because the associated efficiencies are rather low and, in the case of neutrons, very difficult to measure.

Storage rings offer unique opportunities for surrogate-reaction experiments. Indeed, the cooled stored ions have a much better beam quality than non-stored radioactive beams. The beam energy definition is excellent, the beam energy resolution ΔE/E is improved from typically 10^{-3} to 10^{-4}-10^{-5}, and the beam size reduced from several mm to 1 mm diameter [11]. The stored ions interact with ultra-thin, windowless gas-jet targets located inside the ring, for which thicknesses up to about 10^{14} atoms/cm^2 can be obtained for light gases as H_2 and D_2. Contrary to single-pass measurements, the stored ions pass the gas target with a repetition rate of the order of 1 MHz (at 10 A MeV). This revolution frequency leads to an increase of the effective luminosity of about a factor 10^6, which then becomes comparable with the luminosity obtained with a target foil in standard experiments. Moreover, because the electron cooler is passed in each turn, the energy lost in the gas target due to target ionization processes is regained. This means that, in contrast to single-pass experiments, the target is traversed every time with virtually constant beam energy. Last but not least, the possibility to store pure isomeric beams would allow us to study the impact of angular-momentum and parity in a unique and very precise way by comparing the results for the same surrogate reaction carried out with the beam in the ground state and in an isomeric state. Therefore, storage rings appear as the ideal devices for surrogate-reaction studies in inverse kinematics.

However, the storage of ions in a ring is strongly hampered by electron capture and stripping reactions between the revolving ions and the residual gas. For this reason, storage rings are operated in ultra-high vacuum (UHV), namely 10^{-11}-10^{-12} mbar. The very demanding UHV conditions of the ring pose major constraints on the particle detectors used for in-ring experiments.

The decay probability in the outgoing decay channel χ of a nucleus A produced in a two-body reaction $X(y,w)A$ between a heavy projectile X and a light target nucleus y in inverse kinematics can be obtained as:

$$P_\chi(E^*) = \frac{N^{C,\chi}(E^*)}{N^S(E^*)\cdot\varepsilon_\chi(E^*)}. \tag{2}$$

Here $N^S(E^*)$ is the so-called "singles spectrum", i.e. the total number of detected target-like nuclei w as a function of the excitation energy E^* of A. $N^{C,\chi}(E^*)$ is the "coincidence spectrum", corresponding to the number of target-like residues w detected in coincidence with the observable that identifies the decay mode (e.g. a fission fragment or the heavy projectile residue formed after γ or particle emission by A) and ε_χ is the associated detection efficiency. The quantity $N^S(E^*)$ corresponds to the total number of formed A nuclei and $N^{C,\chi}/\varepsilon_\chi$ to the number of A nuclei that have decayed via channel χ. E^* is obtained by measuring the kinetic energy and angle of w and applying energy and linear momentum conservation for the two-body reaction $X(y,w)A$. The kinematics of transfer reactions involving a heavy-ion beam (e.g. ^{238}U) at 9 A MeV and a light target nucleus (e.g. ^{2}H) leads to target-like residues covering a broad range of angles from 0 to 180 degrees, whereas the associated projectile-like residues and the elastic scattered beam are very much forward focused with maximum emission angles of about 1°. The fission fragments are emitted within a cone of typically 20°.

The upper part of Fig. 1 shows the detector arrangement that we want to develop for the simultaneous measurement of the different decay probabilities. Two detectors, one placed upstream and the other downstream the gas-jet target, will be used to identify and measure the kinetic energies and angles of the target-like nuclei. These detectors are followed by a fission detector to detect fission fragments in coincidence with the target-like nuclei and by a heavy-residue detector to detect the projectile-like nuclei produced after γ or particle emission in coincidence with the target-like residues. The latter detector is placed after one of the dipoles of the ring, which will separate the heavy residues from the beam according to their magnetic rigidity. Note that the coincidence with the target-like nuclei will suppress the background caused by the detection of beam-like residues coming from electron capture and stripping reactions. The coincidence will also allow us to discriminate the good events from elastic scattering events in the gas-jet target.

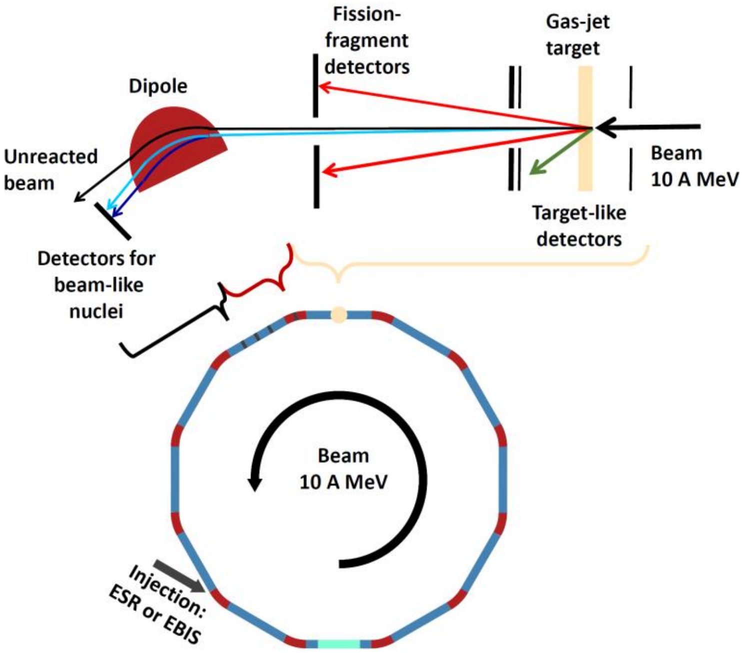

Fig. 1. Schematic view of the complete set-up for indirect neutron cross-section measurements. The CRYRING storage ring is shown in the lower part. The upper part shows the particle detectors that will be placed inside the CRYRING for the simultaneous measurement of fission, γ- and particle-emission probabilities.

Our objective is to detect fission fragments with solar cells, which seem to be a very interesting and cost-efficient alternative to Si detectors, as they are much more resistant against radiation [12]. However, they have never been used in inverse kinematics with fission-fragment energies of about 10 A MeV, and their UHV compatibility has not been demonstrated. We will investigate the energy and time response of solar cells to heavy ions at energies around 10 to 15 A MeV at the GANIL facility in Caen, France, where nuclei in the fission-fragment mass region A=80-140, such as ^{84}Kr and ^{136}Xe isotopes, can be accelerated to these energies. In addition, we have already performed first tests of the UHV compatibility of solar cells. The results are extremely encouraging. Indeed, after baking the solar cells at 160 degrees Centigrade for two weeks, it was possible to reach a pressure of $6{\cdot}10^{-10}$ mbar with just a primary pump and a turbo pump. We estimate the solar cell outgassing rate to about $2.4{\cdot}10^{-13}$ mbar·l/s/cm^2. Moreover, the response of the cells did not deteriorate after the two weeks of bake-out. A complete UHV test bench is being developed at the CENBG to achieve the vacuum requirements of the CRYRING.

In addition, we have performed very detailed detector and ion-optic simulations. We have mainly considered the ^{238}U(d,d’) and ^{238}U(d,p) reactions at 9 A MeV. The simulations show that the efficiencies for detecting the residues of the different decay channels are much larger than in traditional, direct-kinematics measurements and that it is possible to simultaneously measure the decay probabilities of *all* decay channels with an excitation energy resolution ranging from 50 to about 300 keV (standard deviation). We would like to stress that this energy resolution is rather close to what is typically obtained by the CENBG collaboration in direct-kinematics. Obtaining a good excitation-energy resolution is essential since the decay probabilities depend strongly on excitation energy near the fission and particle-separation thresholds.

The so called *β*-Oslo method [13] has been proposed as an indirect method to infer (n,γ) cross sections. In this approach, high-lying levels in the nucleus of interest are populated via *β* decay. A total absorption spectrometer is used to measure gamma-rays and thereby determine the level density, as well as the γ-ray strength function experimentally. These measurements are then combined with theoretical calculations to derive the neutron capture cross section. This method requires normalizing the measured level density to the number of known discrete levels at low excitation energy and to neutron (or proton) resonance data at high excitation energy. However, for very exotic nuclei, these data do not exist. Moreover, contrary to our approach based on the measurement of the probabilities of all open decay channels, the *β*-Oslo method does not give any information on the decay channels that compete with γ emission, i.e. particle emission and fission, which is needed to obtain a reliable result of the neutron capture cross section from the theoretical calculation.

3. Direct measurements of neutron cross sections at storage rings

The direct measurement of neutron cross sections of short-lived nuclei in inverse kinematics requires the interaction of a radioactive beam with a free-neutron target. The neutron is an unstable particle that decays with a half-life of about 10 minutes. Therefore, to produce a free neutron target one needs a source that continuously generates neutrons like a nuclear reactor or a spallation source. However, because of the relatively low beam intensities and neutron target thickness, the luminosity that would be obtained by directly coupling a radioactive-ion source with a neutron target is too low to perform cross-section measurements. As proposed for the first time by René Reifarth and Yuri Litvinov [14], this can be solved by using a storage ring since the revolving frequency of the stored ions leads to a significant increase of the luminosity.

The facility proposed by Reifarth and Litvinov [14], consists in the coupling of an ISOL facility with a storage ring to a high-flux nuclear reactor. The reactor would serve to fill in permanence a small part of the storage ring with a gas of neutrons practically at rest, thus providing a neutron target. The authors showed that neutron densities of $2\cdot10^{10}$ neutrons/cm^2 can be achieved, making possible the measurement of neutron-induced cross sections in inverse kinematics for beams with half lifes of only few minutes or even less. Capture cross sections (n,γ) can be measured using Schottky spectroscopy, and two-neutron emission (n,2n) and charged-particle-emission cross sections (n,p) and (n,α) can be obtained by detecting the beam-like products at the appropriate positions inside the ring. Because of the rather isotropic emission of the fission fragments in the centre of mass reference system, the study of neutron-induced fission with this facility is only possible with a reasonable efficiency for beam energies beyond few A MeV.

In a recent publication we investigated the possibility to replace the very demanding reactor by a specially designed spallation neutron source [15]. The advantages of such a setup over the reactor approach are manifold: No critical assembly is required, and therefore, the safety and security regulations are much less stringent. No actinides at all are used or produced. In particular, no minor actinides are produced avoiding long-lived radioactive waste. Last but not least, there are considerably fewer γ-rays per neutron. The complete facility is sketched in Fig. 2. Neutrons are produced by protons impinging on a tungsten spallation target (brown). The proton beam pipe (red) is orientated perpendicular to the ion beam pipe (light brown). The beam pipes do not intersect. The neutrons produced in the spallation process are moderated in the surrounding heavy water (blue). They penetrate the ion beam pipe and act as a neutron target for the ions. The ion beam pipe is part of a storage ring outside the moderator. The storage ring may contain additional equipment like an electron cooler (green), Schottky pickups and particle detectors (gray).

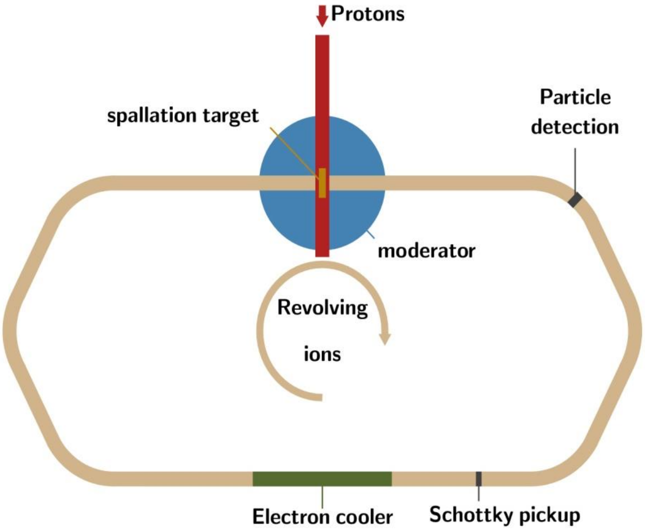

Fig. 2. Sketch of the set-up proposed in [15] for the direct measurement of neutron cross sections in inverse kinematics.

GEANT simulations show that similar neutron densities as in a research reactor can be reached in the ion beam pipe if the spallation target is surrounded by a moderator with heavy water (D_2O) of 2 m radius. More precisely, using a 100 μA proton beam with 800 MeV impinging on a cylindrical Tungsten spallation target with a radius of 1.5 cm and a length of 10 cm, a neutron density of $8 \cdot 10^9$ neutrons/cm^2 can be achieved. Such a proton beam is available at the LANSCE accelerator of the Los Alamos National Laboratory (LANL). LANSCE also has an installation for the production of radioactive beams, which makes the LANL a very interesting option for the development of this facility. Another very promising option is the radioactive-ion-beam facility HIE-ISOLDE at CERN, where it has been proposed to transfer the Test Storage Ring of Heidelberg [11]. The CERN Proton Synchrotron delivers $3 \cdot 10^{12}$ protons/s at 20 GeV whose interaction with a tungsten target with 2.5 cm radius and 50 cm length would lead to a neutron density of $5.4 \cdot 10^8$ neutrons/cm^2 in the ion pipe. Last but not least, the FAIR facility will provide about $5 \cdot 10^{12}$ protons/s with an energy of 28.8 GeV, which together with its variety of specialized instruments like fragment separators and storage rings provides numerous opportunities to realize the facility described here.

4. Conclusions

Storage rings offer unique opportunities for the determination of neutron cross sections of short-lived nuclei, which are essential in several domains such as nuclear astrophysics, nuclear reactor technology and nuclear medicine.

We have presented two future projects. One of the projects will be developed in the following years, it makes use of surrogate reactions to indirectly infer neutron cross sections. It requires an in-ring setup for the measurement of decay probabilities induced by transfer or inelastic-scattering reactions. The set-up consists of two particle detectors surrounding a gas-jet target, a fission-fragment detector and a heavy residue detector located after one of the dipoles of the ring. We are currently investigating the possibility to use solar cells as fission detectors. Detailed ion-optic and detector simulations show that expected detection efficiencies are much larger than in traditional surrogate-reaction experiments in direct kinematics. Moreover, with the proposed set-up it will be possible to measure for the first time simultaneously fission, γ- and particle emission probabilities with an excitation energy resolution of the order of 50 to 300 keV (standard deviation).

The other project will be developed in the next decades. This ambitious project would allow us to directly measure neutron cross sections in inverse kinematics. It is based on the combination of a radioactive-ion beam facility, a storage ring and a spallation neutron source. With such a facility it will be possible to measure (n,γ), (n,2n), (n,p) and (n,α) cross sections for nuclei with half-lives of a few minutes. The LANL, CERN and FAIR appear as very promising options for the development of this future facility as they already have part or the totality of the main required components. (n,f) cross sections can be directly measured for neutron energies above few MeV, only. The fission cross sections at lower neutron energies will be inferred by measuring the fission probabilities associated to surrogate reactions. Therefore, the two described projects are complementary.

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