Indirect measurements of neutron cross-sections at heavy-ion storage rings

A. Henriques[1], B. Jurado[1], M. Grieser[2], D. Denis-Petit[1,3], T. Chiron[1], L. Gaudefroy[3], J. Glorius[4], C. Langer[5], Yu. A. Litvinov[4], L. Mathieu[1], V. Méot[3], R. Pérez-Sánchez[1], J. Pibernat[1], R. Reifarth[6], O. Roig[3], B. Thomas[1], B. A. Thomas[1], J. C. Thomas[7], I. Tsekhanovich[1]

[1] Centre d'Etudes Nucléaires de Bordeaux Gradignan (CENBG), CNRS/IN2P3-Université de Bordeaux, Bordeaux, France
[2] Max-Planck Institut für Kernphysik, Heidelberg, Germany
[3] CEA-DAM DIF, Arpajon, France
[4] GSI Helmholtzzentrum fuer Schwerionenforschung, Darmstadt, Germany
[5] FH Aachen University of Applied Sciences, 52066 Aachen, Germany
[6] University of Frankfurt, Frankfurt, Germany
[7] Grand Accélérateur National d'Ions Lourds (GANIL), CEA/DRF-CNRS/IN2P3, Caen, France

E-mail: henrique@cenbg.in2p3.fr

Abstract

Cross sections for neutron-induced reactions of short-lived nuclei are essential for nuclear astrophysics since these reactions in the stars are responsible for the production of most heavy elements in the universe. These reactions are also key in applied domains like energy production and medicine. Nevertheless, neutron-induced cross-section measurements can be extremely challenging or even impossible to perform due to the radioactivity of the targets involved. Indirect measurements through the surrogate-reaction method can help to overcome these difficulties.

The surrogate-reaction method relies on the use of an alternative reaction that will lead to the formation of the same excited nucleus as in the neutron-induced reaction of interest. The decay probabilities (for fission, neutron and gamma-ray emission) of the nucleus produced via the surrogate reaction allow one to constrain models and the prediction of the desired neutron cross sections.

We propose to perform surrogate reaction measurements in inverse kinematics at heavy-ion storage rings, in particular at the CRYRING@ESR of the GSI/FAIR facility. We present the conceptual idea of the most promising setup to measure for the first time simultaneously the fission, neutron and gamma-ray emission probabilities. The results of the first simulations considering the ^{238}U(d,d') reaction are shown, as well as new technical developments that are being carried out towards this set-up.

Keywords: neutron-induced reactions, surrogate reactions, inverse kinematics, heavy-ion storage rings

1. Introduction

The synthesis of elements from iron to uranium that takes place in stars, can only be understood through the study of neutron reactions on radioactive nuclei [1,2]. Neutron cross sections of radioactive nuclei are also important for industrial purposes, e.g. for more sustainable and efficient energy production and in the search for new therapeutic radionuclides for medical diagnostic and treatment.

It is known that heavy elements are produced via neutron induced reactions and depending on the neutron density, we can have the slow (s) and the rapid (r) neutron-capture process. Each of the processes produces about half of the heavy elements. In the s-process few neutrons are available, unstable isotopes will more likely decay than capture a neutron and the nucleosynthesis path takes place only one or two neutrons away from the β stability. As for the r-process, the neutron
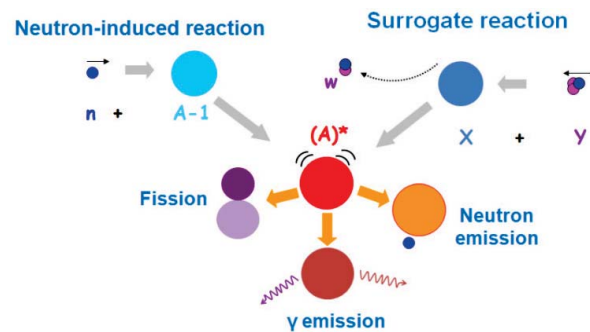

Figure 1 - On the left side is the formation of the compound nucleus A from a neutron induced reaction. On the right side of the picture is shown the formation of the same compound nucleus using an alternative reaction, a surrogate reaction. At low energies (En<5 MeV) the compound nucleus can decay by fission, γ- and particle-emission.*

density is very high, many neutrons will be captured consecutively and form very heavy isotopes of a certain element before they decay.

At the end of the r-process path one can find extremely neutron-rich nuclei in the actinide region. These nuclei will most likely fission. The fission fragments will then feed the distribution of mid-mass range nuclei. Neutron capture will continue forming new heavy elements that will fission. This will form the so-called fission recycling process and after few cycles the abundances may become dominated by the fission-fragment distributions. Fission cross-sections, yields and fission barriers play a key role in understanding the r-process.

Direct measurements of neutron-induced reactions on unstable isotopes are very difficult for several reasons. Capture measurements require the detection of γ-rays in the presence of a γ-ray background due to the decay of the target material [3]. The fission measurements face the background from α-emission or spontaneous fission competing with the signals from the neutron-induced fission. The radiation from the radioactive sample can severely damage the detectors [4] and finally the production and handling of radioactive samples is severely restrained by health and safety regulations.

1.1 Surrogate reactions

The most promising approach to overcome the difficulties of measuring neutron induced reactions is to use the surrogate reaction method. Via the surrogate reaction, the compound nucleus of interest is produced by using an alternative reaction to the neutron induced reaction, as shown in figure 1.

The excited compound nucleus after being formed can decay by γ-emission, neutron-emission or fission, like in the neutron induced reaction. The decay probabilities of the compound nucleus can be then used to tune model parameters that will lead to much more accurate predictions of the desired neutron cross sections. Surrogate reactions of interest are for example transfer or inelastic scattering reactions with light nuclei, e.g. (d,p) or (d,d'). The measurement of decay probabilities requires: a) the identification of the emitted light particle, b) to precisely measure its kinetic energy and emission angle in order to determine the excitation level in the compound-nucleus, and c) the detection of the products of the compound nucleus decay.

The power of the surrogate method has been proven very successfully at high excitation energies for neutron induced fission in regular kinematics with ^{1}H, ^{2}H, ^{3}He and ^{4}He beams [5-7], and very recently also for neutron radioactive capture [8-10]. Since 2000, the CENBG, together with other laboratories in France and abroad, has been performing experiments to study the surrogate reaction method [5,7]. In the past years, the experimental set-up was improved in order to simultaneously measure fission and γ-emission probabilities [11].

The principle of the experiments carried out so far is illustrated in figure 2, a light particle beam is directed onto the target. After the reaction, the heavy target nucleus is left in an excited state. The ejectiles are detected with position-sensitive Si telescopes located at backward angles, the fission fragments with solar cells and γ-rays with C_6D_6 liquid scintillators. The exclusion of events where the origin of the γ-ray is a fission fragment was successfully achieved [10,12].

Although very promising, the studies done up to now are not sufficient to establish surrogate reactions as a standard tool to infer neutron cross sections in regions where no data exist. One of the main issues is that the surrogate reaction can lead to compound nucleus angular momentum and parity distributions very different from the ones populated in the neutron-induced reaction. In particular, compound nuclei located near shell closures are expected to be very sensitive to spin-parity effects. For this reason systematic studies throughout the nuclear chart are in demand.

From a technical point of view, the study of surrogate reactions in direct kinematics faces several limitations: unavailability of targets from short-lived nuclei; high background from the competing reactions with the target contaminants and backing; and low detection efficiencies. Regarding the last point, the heavy products resulting from the the decay of the compound nucleus are stopped in the target and cannot be detected with particle detectors. Therefore, the measurement of γ and neutron-emission probabilities in direct kinematics relies in the detection of the γ-rays or the neutrons emitted by the compound nucleus, which is very difficult due to the very low detection efficiencies.

Inverting the kinematics of the reaction means accelerating the heavy nucleus while keeping at rest the light particle (protons, deuterons, α-particles...) which act as a target. The inverse kinematics technique gives access to short-lived nuclei and allows to directly detect the heavy products of the compound nucleus after γ and neutron-emission.

There are several radioactive-ion-beam facilities world-wide following this approach. Nevertheless, experiments have to be designed carefully in order to have a high reaction probability. For example, when considering surrogate reactions the most promising isotopes are gases (H and He), but high areal densities of such molecules are difficult to reach. Several issues associated with the target (size, thickness, support and backing) result in the loss of resolution of the excitation energy of the compound nucleus. Such difficulties can be overcome if the surrogate experiments are performed at storage rings.

1.2 Surrogate reactions at Storage Rings.

A heavy-ion storage ring is an ensemble of beam pipes and electro-magnetic devices (dipoles, quadrupoles, etc.) arranged in a closed geometry where the heavy ions turn with high frequencies, about 1 MHz at 10 MeV/u. The storage of heavy ions requires minimizing atomic reactions between the stored beam and the residual gas inside the ring. Therefore, heavy-ion storage rings are operated at Ultra-High Vacuum (UHV) conditions (10^{-11} to 10^{-12} mbar) [13].

The most important capability of a storage ring is beam cooling, which allows the reduction of the energy and position spread of the stored ions induced by the reaction mechanism used to produce the ions or by the interaction with targets. Beam cooling takes typically seconds and sets the lower limit of the half-live of the radioactive ions that can be prepared. The combination of the electron cooler and the dipole magnets ensures an extraordinary quality of the stored beam. In addition, the electron cooler can compensate the energy loss of the beam in the target. Hence, the ions pass the target always at the same energy. Moreover, the frequent passing in the reaction zone allows ultra-thin gas-jet targets (10^{13} atoms/cm^2) to be used and therefore no windows are necessary. The beam will only interact with the desired material and in a well-defined interaction zone.

The UHV environment poses severe constraints to in-ring detection systems because the vacuum conditions have to be improved by several orders of magnitude compared to regular experiments. For this reason nuclear reactions have started to be measured only very recently at the Experimental Storage Ring (ESR) of the GSI/FAIR facility in Darmstadt, Germany [14]. These pioneering experiments open up unique and yet unexplored possibilities to measure decay-probabilities induced by surrogate reactions in inverse kinematics.

2. Decay probabilities measurements at heavy-ion storage rings

The outstanding beam quality of heavy-ion storage rings, along with the electron cooler enables a very precise measurement of the excitation energy of the decaying nucleus.
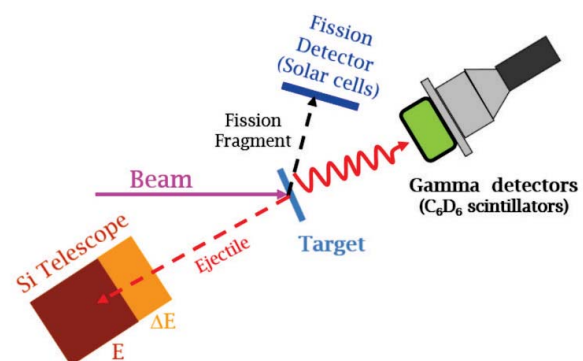

Figure 2 - Schematic view of the experimental set-up for the simultaneous measurement of fission and γ-emission probabilities induced by surrogate reactions in direct kinematics.

Another very interesting possibility of the rings is the study of isomers. For example, if we consider the case of an isomer with a lifetime significantly larger than the ground state. The ground state will decay faster than the isomer. After the cooling, the beam will only contain isomers. Isomers have a very different configuration (angular momentum, parity) than the ground state and offer therefore a unique possibility to investigate the influence of angular momentum on the decay probabilities.

The promising advantages of performing surrogate reaction experiments in inverse kinematics using heavy-ion storage rings led to the study of the feasibility of this idea proposed by B. Jurado [7]. The future surrogate measurements are proposed to be carried out at the CRYRING@ESR [15] storage ring of GSI/FAIR and a Technical Design Report (TDR) describing the details of the required set-up will be submitted in the near future to GSI/FAIR.

A first experiment considering a ^{238}U beam and a D_2 target will allow us to have access to the $^{238}U(d,p)$ reaction for which there is neutron data (n+^{238}U) available to compare. Thus, allowing us to test the theoretical tools used to correct for the spin/parity mismatch.

2.1 The future setup

The future set-up aims at simultaneous measurements of fission, γ- and particle- emission probabilities which has never been achieved before. The set-up is presented in figure 3, where a schematic view of the CRYRING is shown on the bottom and on the top is a zoomed view of the required detection system.

The set-up consists of three main detection systems: a target-like particle detector, a fission fragment detector and beam-like residue detector located after the two dipoles. The latter detector will allow to detect the heavy residues after neutron and γ-emission.
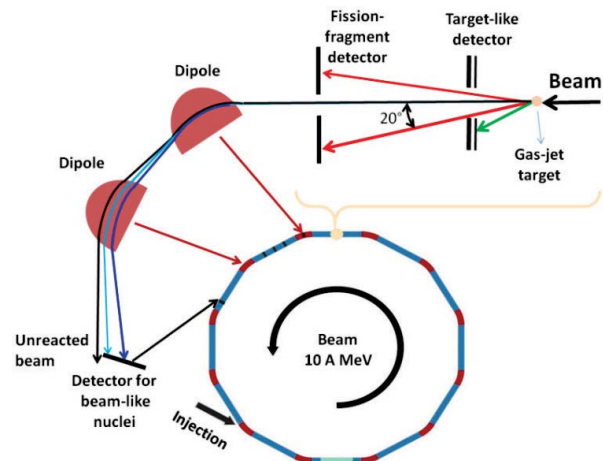
Figure 3 - The upper part shows a schematic view of the experimental setup that will be placed inside the CRYRING for the simultaneous measurement of fission, γ- and neutron-emission probabilities. The trajectories of the heavy beam-like residues produced after γ- and neutron emission are shown in light blue and blue, respectively.

Just after the target, two particle telescopes ΔE-E (DSSSD-SiLi) will be placed to identify and measure the kinetic energies and angles of the light target-like nuclei ejected during the surrogate reaction. These detectors will be placed 4 cm away from the target and inside a movable pocket with a 25 μm stainless steel window. Downstream of the target, covering forward angles, a detection system, placed directly in UHV, composed of solar cells will be used to detect fission fragments in coincidence with the target-like detectors. The beam-like residues formed after γ-ray or particle emission of the compound nucleus, with almost the same direction and energy as the incoming beam will continue down the ring, will be deflected according to their magnetic rigidity by the two dipole magnets and be separated and detected at the focal plane. Thanks to inverse kinematics, fission fragments and beam-like products are emitted in forward direction, which results in detection efficiencies that are much larger than in the traditional direct kinematics measurements.

The beam-like products are measured in coincidence with the target-like ejectiles. The described set-up will allow to measure the decay probabilities, P_χ, considering the ratio between the coincidence events, $N_{coinc,\chi}$, and the number of target-like nuclei, N_{targ}. The number of coincidences has to be corrected by the corresponding efficiency, ε_χ, of the decay channel. The decay probabilities, P_χ, are given by:

$$P_\chi = \frac{N_{coinc,\chi}(E^*)}{N_{targ}(E^*) \cdot \varepsilon_\chi}$$

Finally, one or two high-purity germanium detectors will be placed close to the target. Such detectors will be used to detect the x-ray signature of the radiative electron capture process and be used to determine the luminosity in the ring which in its turn gives access to determine the cross sections as a function of angle of the target-like nuclei.

2.2 Simulations

The physical feasibility of the proposed set-up started to be evaluated by performing Geant4 simulations using the G4beamline particle tracking program [16]. The first simulations considered the inelastic scattering $^{238}U(d,d')$ reaction at 11 MeV/u and were used to evaluate the excitation energy resolution, the foreseen detection efficiency and the transmission efficiency of the beam-like residues through the ring up to the detector for beam-like nuclei.

The simulations considered all the residues of the ^{238}U(d,d') reaction performed in inverse kinematics, tracking them from the target to the different detection systems. For this the reaction kinematics was coded in an event generator. This generator can be described in two steps: the compound nucleus formation and its decay by fission, neutron and γ-emission. Several conditions, including in-ring measurement conditions, were accounted for: a) the ^{238}U beam at 11 MeV/u with a momentum spread ($\frac{\Delta P}{P}$) of 2x10^{-4}, a beam emittance (phase space area) ranging from 0.05 to 0.5 mm·mrad and an horizontal dispersion at the target position of 1.5 m. b) the deuteron target of cylindrical shape with a 1 mm diameter c) the scattered deuterium emitted at 40° with respect to the beam axis and d) a compound nucleus excitation energy of 8.2 MeV (about 2 MeV above the neutron separation energy of ^{238}U).

At a first instance, we studied the expected excitation energy resolution. This considered the previous conditions and the tracking of the scattered deuterons into the telescopes placed behind a 25 µm stainless steel foil. The segmentation of the telescopes and a detector energy resolution of 1% were taken into account. Figure 4 shows the reconstructed excitation energy which is given by:

$$E^* = E_{238_U} + Q - E_{2_H} - \frac{1}{M_{238_U}}(M_{238_U}E_{238_U} + M_{2_H}E_{2_H} - \sqrt{M_{238_U}E_{238_U}M_{2_H}E_{2_H}}\cos\theta)$$

Where E stands for energy, M for mass and θ is the polar angle of the target-like particle. From the simulations it was observed that the telescope resolution was the most limiting factor to the excitation energy resolution. The segmentation of the detector and stainless steel window effects were almost negligible.

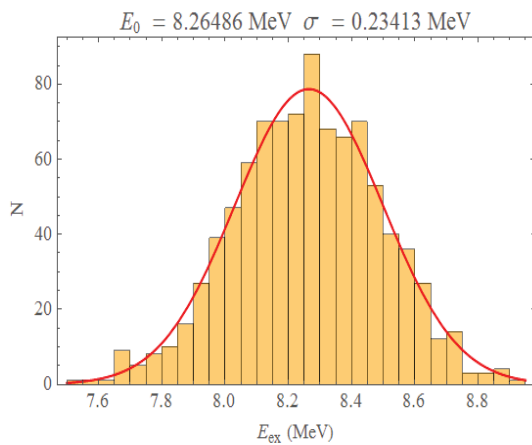The fission process of ^{238}U was simulated using the GEF code [17]. GEF provides the atomic mass, number and energy of the two fission fragments. The kinematics of the reaction was also taken into account, including the anisotropy of the angular distribution of the fission fragments in the center of mass reference system. The latter distribution can be described by the function $1 + \alpha \cdot cos^2\theta_{ff}^{cm}$, where α is the anisotropy factor and θ_{ff}^{cm} the fission fragments angle in the centre of mass frame. Figure 5 show the expected polar-angle distribution of the light and heavy fragment in the laboratory frame.

It is necessary to leave a gap of 10 cm by 4 cm in the fission detector to let the beam and beam-like residues continue their path. Considering this fact and the fragments distribution, it was possible to optimize the geometric effiency of the detector. At a distance of 60 cm from the target we obtain a maximum detection efficiency of 96%.

In the case that the excited compound nucleus ^{238}U decays by neutron (into ^{237}U) or γ-emission (into ^{238}U), the beam-like residues will continue with a trajectory very close to the beam.

Figure 4 - Compound nucleus excitation energy reconstruction obtained from the simulation of the ^{238}U(d,d') reaction. The standard deviation of 234 keV in the excitation energy is mainly due to the detector resolution. The beam emittance considered was 0.05 mm·mrad.

These decays were simulated: a) the γ decay considered one single gamma equal to the excitation energy of the compound nucleus. b) The emitted neutron energy was sampled up to the maximum neutron kinetic energy possible. The corresponding particle phase spaces were tracked and the position of each residue at the focal plane after the second dipole downstream of the target is shown in figure 6.

The transmission efficiency along the storage ring beam line up to the focal point is of 100% for the beam-like ^{238}U residues and of 98.8% for the ^{237}U residues. This shows the power of performing such experiments in inverse kinematics where we can expect a detection efficiencies close to 100%, much larger than the ones obtained in direct kinematics.
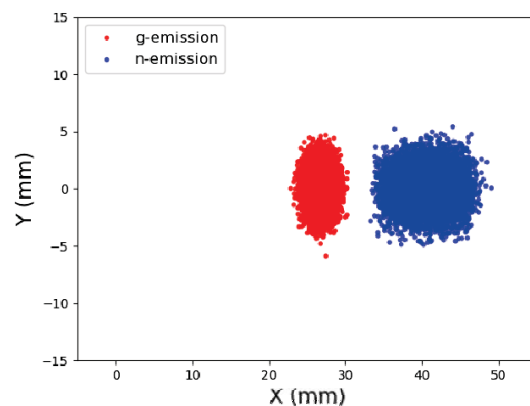
2.3 Solar Cells

Figure 6 - Simulated beam-like residues position after gamma (red) and neutron (blue) emission at the focal point. The beam emittance considered was 0·5 mm·mrad.

In the previously presented setup, we propose to use solar cells as heavy ion detectors, in particular to detect the fission fragments. The common solar cells found at the rooftops have been used to detect heavy ions at low energies of about 1 MeV/u for several decades [18-20]. They are considered a good and cost-efficient alternative to Si detectors, due to their radiation resistance properties [20], making solar cells an attractive detection system.

We have started to conduct several tests to assess the feasibility to use solar cells as heavy ion detectors by studying their response to heavy ion beams of about 10 MeV/u and their compatibility with the extreme high vacuum required in the storage rings and by developing new pre-amplifiers suited to our specific needs.

The first exploratory irradiations of solar cells to heavy ions with energies above 1 MeV/u took place in May 2018 at the GANIL facility, France. Ilustrative energy and time spectra are shown in figure 7 for a 10x10 mm^2 cell irradiated with a ^{129}Xe beam at 10 MeV/u.

Up to now, we have used ^{84}Kr and ^{129}Xe beams, with energies ranging from 2 to 15 MeV/u, to study the response of solar cells. The energy resolution (standard deviation) for a 10x10 mm^2 cell was found to be between 2-3% while the time resolution (FWHM) was of about 4 ns.
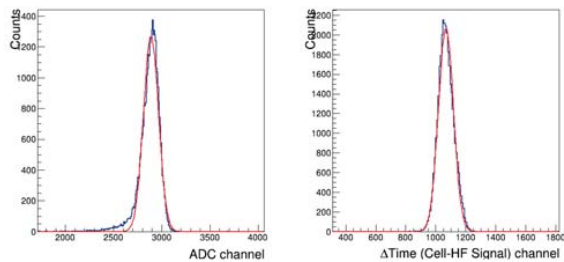

Figure 7 - Energy (left) and time (right) spectra of a 10x10 mm^2 cell irradiated with a ^{129}Xe beam at 10 MeV/u.

The outgassing rate of solar cells and detector supports is also being investigated at the CENBG, where with the aid of a residual gas analyser, we will determine if the material outgassing is in agreement with the in-ring requirements. A preliminary measurement performed at the vacuum laboratory of GSI showed a very low outgassing, bellow 10^{-11} mbar·l/(s·cm^2) after baking for 48 hours at 200°C.

The available pre-amplifiers were not adapted to our needs and, in addition, their technology was outdated. The development of new pre-amplifiers has also been carried out by J. Pibernat at the CENBG. The challenge in the development of the pre-amplifiers resides on the large capacitance of the cells, about 40 nF/cm^2. Up to now the solar cell electronic model has been validated and this allowed to develop new pre-amplifiers that were successfully tested at the GANIL facility during 2019 using ^{238}U beam at 3.8 MeV/u [21]. The solar cell electronic model consists of one capacitor (Cd=38 nF/cm^2) in parallel with a resistor (Rp=5 kΩ) which are in their turn connected in series with the resistor Rs (0.1-10 Ω). The new pre-amplifiers are already able to achieve the same signal-to-noise ratio and response as the old ones.

Conclusions

Surrogate reactions are the most promising method to infer neutron-induced cross-sections of short-lived nuclei, in particular to tune model parameters in regions of the nuclear chart where no data is available. Still, systematic studies are needed to fully understand surrogate reactions. To pursue these systematic studies, we propose to measure decay probabilities in inverse kinematics at storage rings. The proposed setup will not only allow to overcome many of the issues found in direct kinematics experiments, it will additionally permit to measure simultaneously, for the first time, fission, neutron and γ-decay probabilities with very high precision data.

The feasibility studies performed so far, via simulations, show the power of such setup as the excitation energy resolution is adequate to study the abrupt change of the probabilities with the excitation energy. Moreover, the expected detection efficiencies of the beam-like residues are much larger than the ones possible in direct kinematics.

Additional studies towards the use of solar cells as heavy ion detectors have already started. All the results are, up to now, positive to include these detectors inside storage rings and detect heavy ions at 10 MeV/u.

Acknowledgements

References

[1] E. M. Burbidge et al., Synthesis of the Elements in Stars. Rev Mod. Phys. **29** 547 (1957)
[2] H. Schatz, Trends in Nuclear Astrophysics. J. Phys. G: Nucl. Part. Phys. **43**, 064001 (2016)
[3] A. Couture, et al, Atomic Data and Nuclear Data Tables **93** (2007)

[4] P. Marini, et al. Phys. Rev. C **96** 054604 (2017)

[5] G. Kessedjian, et al., Phys Lett. B **692**, 297 (2010)

[6] J.E. Escher, et al., Reviews of Modern Physics 84 (2012)

[7] Jurado, B., et al., European Phys. J.l Web of Conf. **146** (2017)

[8] J.E. Escher, et al., Physical Review Letters **121**, 052501 (2018)

[9] A. Ratkiewicz, et al., Phys. Rev. Lett. **122**, 052502 (2019)

[10]   R. Pérez Sánchez, PhD thesis, Universite de Bordeaux (2019)

[11]   R. Pérez Sánchez, et al., NIM A **933**, 63 (2019)

[12]   Q. Ducasse, et al., NIM A **826** (2015)

[13]   Yu.A. Litvinov, NIM B 317 (2013) 603–616

[14]   J. Glorius et al., Phys Rev Lett **122**, 092701 (2019)

[15]   M. Lestinsky, et al., Eur. Phys. J. Special Topics **225** (2016)

[16]   http://www.muonsinternal.com/muons3/G4beamline

[17]   K.-H. Schmidt, et al., Nuclear Data Sheets **131** (2016)

[18]   S. Siegert, et al., NIM **164** (1979)

[19]   M. Hsieh, et al, IEEE Electron Device Letters **4** (1981)

[20]   N. Ajitanand, et al., NIM A **300** (1991)

[21]   A. Henriques, et al., submitted to NIM A (2019)