Neutron-capture rates for explosive nucleosynthesis: the case of $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$

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Neutron-capture reactions play an important role in heavy element nucleosynthesis, since they are the driving force for the two processes that create the vast majority of the heavy elements. When a neutron capture occurs on a short-lived nucleus, it is extremely challenging to study the reaction directly and therefore the use of indirect techniques is essential. The present work reports on such an indirect measurement that provides strong constraints on the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction rate. Before the present results, this reaction was unconstrained and the purely theoretical reaction rate was highly uncertain. The new uncertainty on the reaction rate based on the present experiment is approximately a factor of 3. The commonly used reaction libraries JINA-REACLIB and BRUSLIB are in relatively good agreement with the experimental rate. The impact of the new rate on weak r-process calculations is discussed.

I. INTRODUCTION

The creation of elements in the universe is one of the major open questions of our time. Multiple fields of science come together and provide different pieces to the nucleosynthesis puzzle. Astronomical observations, meteoritic analysis, astrophysical simulations, nuclear structure and reactions calculations, and experimental nuclear data are all paramount for the better understanding of these complex astrophysical processes. Since the original publications of a comprehensive picture of stellar nucleosynthesis 60 years ago,[1][2], the field has evolved and expanded. Many questions have certainly been answered, however many still remain, and new ones have been formulated. New techniques and new tools are developed constantly, each of which opens a new window into our understanding of the universe. For example, the recent observation of gravitational waves is such a “window”, with great potential to enrich our knowledge of compact objects during merging events[3].

One particularly challenging piece of the stellar nucleosynthesis puzzle is the origin of the heavy elements (heavier than Fe). Three processes are considered responsible for creating the bulk of the isotopes of these elements: the slow neutron capture process (s process)[4], the rapid neutron capture process (r process)[5][6], and the p process[7][8], which is responsible for producing the small fraction of neutron-deficient isotopes. The two neutron-induced processes dominate the isotope production, with the s process occurring over longer time scales ($\sim 10^3$ years) and progressing along the valley of stability, and the r process extending far into exotic neutron-rich nuclei during extremely short time scales ($\sim 1$ s). The explosive nature of the r process and the fact that the nuclei involved are very neutron-rich and largely inaccessible for experiments at current facilities, are two of the many reasons that keep the r process at the center of attention, despite six decades of study.

The astrophysical site where the r process occurs has not been unambiguously determined. Many scenarios have been proposed over the years (e.g. reviews[5][6]), with the most dominant so far being the core-collapse supernovae, e.g. [9][12], and the neutron-star merger, e.g. [13][15], scenarios. The latter scenario has recently gained even more interest because of the possibility of a “kilonova” afterglow coming from the decaying r-process elements[16]. Recent observations of dwarf spheroidal galaxies[17] showed that the observed patterns are consistent with an r process taking place in rare events, consistent with the neutron-star merger scenario but not exclusively. In addition, there is the possibility for contributions from more than one astrophysical scenario. In such a case, heavy elements (A $> 120$) are considered to be produced in a “main” r process, while the lighter elements with A $\approx 80 – 120$ may have strong additional contributions or even a distinct origin (“weak” r process). It is
therefore clear that the study of the r process is a very lively field with a continuous flow of improved astrophysical scenarios and new observational information.

On top of the large effort to understand the uncertainties associated with the astrophysical conditions, it is extremely important to provide accurate nuclear physics input. Sensitivity studies show that the r-process abundance distribution changes significantly when varying the nuclear physics input within uncertainties (e.g. most recently [18–20] and references therein). The properties that influence the r-process flow and the final abundance distribution are nuclear masses, β-decay rates, β-delayed neutron emission probabilities, and neutron-capture reaction rates. Experiments have provided significant information for masses and β-decay properties, and new experiments are being planned at current and future facilities. However, neutron-capture reaction rate measurements have been extremely challenging experimentally due to the fact that neither of the two reactants (neutron and short-lived neutron-rich nucleus) can be developed as a target for accelerator based measurements. For this reason the development of indirect techniques that will provide experimental constraints on these important reactions is critical. Several techniques have been developed or are currently under development, such as the surrogate reaction technique [21–23], the γ-ray strength function method [24], and the Oslo method [25, 26]. These techniques are all reaction-based, and as such, they require a relatively high beam intensity to be applied. An alternative technique, the β-Oslo method, was proposed recently, which is a combination of the traditional Oslo method with β decay and total absorption spectroscopy techniques [27]. This latter technique is the focus of the present work, applied for the first time for the β decay of 69Co, populating 69Ni and extracting the reaction rate of the 68Ni(n,γ)69Ni reaction. This reaction has been identified in a weak r-process sensitivity study [18] as one of the reactions that influences the final abundance distribution.

Fig. 1 shows a fraction of the chart of nuclei around the region of interest. The color code represents the variation in the predicted reaction rate using the code TALYS-1.6 [29] with different nuclear reaction input parameters (ratio between maximum and minimum rate at 1.5 GK). Specifically, a compound nucleus neutron-capture reaction cross section is calculated using the relevant nuclear masses, the neutron-nucleus optical model potential (nOMP), the nuclear level density (NLD), and the γ-ray strength function (γSF). For the nuclei shown in Fig. 1 the majority of the masses are known experimentally and are listed in the Atomic Mass Evaluation 2012 (AME2012). Nuclei marked with a black dot do not have a mass listed in AME2012, or their mass is the result of an extrapolation. The neutron-capture reaction rate calculations seem to change with the nOMP variation only mildly [30]. Therefore, the two parameters that dominate the uncertainties in the reaction rates are the NLD and the γSF. These are the two quantities that were varied to create Fig. 1. Table I presents a list of the NLD and γSF models used in these calculations. For the reasons discussed in [28], out of the six available NLD models in TALYS-1.6, the temperature-dependent Hartree-Fock-Bogoliubov level densities using the Gogny force based on Hilaire’s combinatorial tables [31] was excluded (ldmodel 6), and out of the five γSF models, the Brink-Axel single Lorentzian formula [32, 33] was also excluded (strength 2).

Fig. 1 shows that the calculations diverge as one moves farther from the last stable isotope. While the variation between models is less than a factor of 5 around stability, the addition of two neutrons (e.g. Mn, Fe and Co chains) results in...
over a factor of 20 difference between models. In particular, for the reaction of interest for the present work, \(^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}\), the calculations exhibit a variation in the theoretical calculations of a factor 35. In the present work we report on the first experimental constraint for this reaction. We use the \(\beta\)-Oslo method to extract the NLD and \(\gamma\)SF for the compound nucleus \(^{69}\text{Ni}\). These two experimental quantities are used as input in the TALYS calculations to provide an experimentally-constrained reaction rate. The experimental results are also used in weak \(r\)-process calculations to show the impact on the final abundance distribution.

### TABLE I: Nuclear level density and \(\gamma\)-ray strength function models used as input in the TALYS calculations presented in Fig. 1

<table>
<thead>
<tr>
<th>Nuclear Level Density Models</th>
<th>TALYS-1.6</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Constant temperature matched to the Fermi gas model (CT + BSFG)</td>
<td>1</td>
<td>[34]</td>
</tr>
<tr>
<td>Back-shifted Fermi gas model (BSFG)</td>
<td>2</td>
<td>[35, 35]</td>
</tr>
<tr>
<td>Generalized super fluid model (GSM)</td>
<td>3</td>
<td>[36, 37]</td>
</tr>
<tr>
<td>Hartree-Fock using Skyrme force (HFS)</td>
<td>4</td>
<td>[38]</td>
</tr>
<tr>
<td>Hartree-Fock-Bogoliubov (Skyrme force) + combinatorial method (HFBS - C)</td>
<td>5</td>
<td>[39]</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>(\gamma)-ray Strength Function models</th>
<th>TALYS-1.6</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kopecky-Uhl generalized Lorentzian (KU)</td>
<td>1</td>
<td>[40]</td>
</tr>
<tr>
<td>Hartree-Fock BCS + QRPA (HF-BCS + QRPA)</td>
<td>3</td>
<td>[41]</td>
</tr>
<tr>
<td>Hartree-Fock-Bogoliubov + QRPA (HF + QRPA)</td>
<td>4</td>
<td>[42]</td>
</tr>
<tr>
<td>Modified Lorentzian (Gor-ML)</td>
<td>5</td>
<td>[43]</td>
</tr>
</tbody>
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### II. METHODS

The experiment took place at the National Superconducting Cyclotron Laboratory, at Michigan State University. A \(^{86}\text{Kr}\) primary beam was accelerated to 140 MeV/nucleon at the Coupled Cyclotron Facility, and was impinged on a Be production target. The A1900 fragment separator [44] was used to select the isotope of interest \(^{69}\text{Co}\) as part of a “cocktail” consisting of approximately twelve isotopes in this mass region [28]. \(^{69}\text{Co}\) was identified event-by-event based on energy loss and time-of-flight information from a plastic scintillator at the focal plane of the A1900, and two Si PIN detectors upstream of the implantation point. The beam ions were implanted in a 1-mm thick double sided silicon strip detector (DSSD), consisting of two sets of sixteen strips, perpendicular to each other, 1.2 mm apart. The signal from the DSSD was fed into dual gain preamplifiers, which provided information about implanted ions (low gain) and the emitted \(\beta\)-decay electrons (high gain). Downstream of the DSSD, a 0.5 mm thick Si detector was used to “veto” any unwanted light ions that were part of the beam cocktail. The \(\beta\) and ion information were correlated in space and time, in order to assign the detected \(\beta\) particles to the appropriate ion. This technique is not free of background, and for this reason random correlations were also created by creating events where \(\beta\) particles and ions were not correlated in space.

The correlation time between ion implantation and \(\beta\) detection was used to confirm the previously known half life of \(^{69}\text{Co}\), as shown in Fig. 2. The best fit to the data is represented by the solid line and it included the exponential decay curve as well as the random background. The extracted half-life of 216(15) ms is in excellent agreement with previous measurements [45–49]. The half life of the daughter nucleus (\(^{69}\text{Ni}\)) is more than a factor of 10 longer than the \(^{69}\text{Co}\) one (ground state 11.2 s, 321 keV isomeric state 3.5 s) and did not have a significant contribution to the fit.

Surrounding the implantation point was the Summing NaI detector (SuN) [50]. SuN is a cylindrical NaI(Tl) scintillator, 16-inches in height and 16-inches in diameter, with a 1.7-inch borehole along its axis. SuN is segmented into eight, optically isolated segments, four above and four below the beam axis. The DSSD implantation detector was placed inside the borehole at the geometric center of SuN. For \(\gamma\) rays emitted at the center of SuN the angular coverage is approximately 98% and the efficiency for single 1-MeV \(\gamma\) full-energy detection is \(\approx 85\%\). Details about SuN’s characterization can be found in [50]. The application of the \(\beta\)-Oslo technique using the SuN detector was presented in two previous publications of our group [27, 28], therefore only a short description of the method will be presented here.

The \(\beta\)-Oslo method relies on the population of the compound nucleus of interest using \(\beta\) decay. Two main pieces of experimental information are required, namely, the populated excitation energy, \(E_x\), and the \(\gamma\)-ray energy, \(E_\gamma\). \(E_x\) can be extracted from SuN by summing all \(\gamma\)-ray energy in an event, following the principle of the technique of total absorption spectroscopy. The segmentation of SuN provides the individual \(\gamma\)-ray energy information, \(E_\gamma\), by treating each segment as an individual detector.
Fig. 3(a) shows the raw $E_x - E_\gamma$ matrix for $^{69}$Ni populated in the $\beta$ decay of $^{69}$Co. This matrix was created by requiring that the implanted ion was identified as $^{69}$Co, a $\beta$ particle was detected in the same pixel of the DSSD within 800 ms of the ion implantation, and no signal was recorded in the Si veto detector. In addition, random correlation background was also subtracted. It is worth mentioning that the experimental matrix shows significant population at excitation energies above the neutron separation energy ($S_n(^{69}\text{Ni}) = 4.5$ MeV). This effect was also observed in the decay of the neighboring nucleus $^{70}$Co [51], and will be explored in more detail for the case of $^{69}$Co in a future publication.

Starting with the raw matrix, the $\beta$-Oslo method follows four main steps:

- Unfolding of the $\gamma$ spectra for each initial excitation energy [52].
- Isolation of the primary $\gamma$-ray spectrum, i.e., the distribution of the first emitted $\gamma$ rays in all the $\gamma$-decay cascades for each initial excitation energy [25].
- Extraction of the functional form of the NLD and the $\gamma$-ray transmission coefficient from the primary $\gamma$-ray spectrum [26].
- Normalization of the NLD and $\gamma$SF [26, 53, 54].

III. RESULTS AND DISCUSSION

In the present work, the unfolding was done based on the response function of the SuN detector. The response function was created using the GEANT4 simulation package [55, 56], which included a detailed description of the experimental setup, and which was validated in detail against experimental data from various radioactive sources and known resonances. The result of the unfolding procedure is shown in Fig. 3(b). For the second step, the well developed and characterized method of Ref. [25] was used to extract the primary $\gamma$-ray distribution per excitation energy. This so called “first generation” matrix is shown in Fig. 3(c). The primary $\gamma$-ray spectrum corresponds to the relative probability of a decay with $\gamma$-ray energy $E_\gamma$ from an initial excitation energy $E_x$, and depend on the NLD at the final excitation energy $\rho(E_x - E_\gamma)$, and the $\gamma$-ray transmission coefficient $\mathcal{T}(E_\gamma)$ [26]:

$$ P(E_\gamma, E_x) \propto \rho(E_x - E_\gamma) \cdot \mathcal{T}(E_\gamma), $$  \hspace{1cm} (1)

where $P(E_\gamma, E_x)$ is the experimental primary $\gamma$-ray matrix. Using Eq. (1), an iterative extraction procedure [26] was applied to obtain the NLD and $\gamma$SF, from the data within $E_{\gamma,\text{min}} = 1.0$ MeV, and $E_x \in [2.2, 5.4]$ MeV. Therefore, the third step results in the extraction of the functional form of the NLD and the $\gamma$SF ($f(E_\gamma)$), where for dipole transitions the latter quantity is connected to the $\mathcal{T}(E_\gamma)$ through:

$$ f(E_\gamma) = \frac{\mathcal{T}(E_\gamma)}{2\pi E_\gamma^2}. $$  \hspace{1cm} (2)

![Graph](image-url)

FIG. 2: (Color online) $^{69}$Co half-life extraction. The left-hand figure shows the experimental data in the filled dots and the best fit in the solid line. The open circles show the random correlation background. The resulting half life of 216(15) ms is shown in the right-hand figure as a red square, compared with black points that correspond to the literature values from Bernas et. al. (1991), Mueller et. al. (1999), Sorlin et. al. (1999), Gaufdefroy (2005), and Daugas et. al. (2011) [45–49].
The fourth step in the analysis procedure is the normalization of the extracted NLD and $\gamma$SF to known quantities. The NLD was normalized to the experimental level density of the low-lying discrete levels of $^{69}$Ni, and the level density at the neutron-separation energy, $\rho(S_n)$. The low-lying level scheme was taken from ENSDF \[48\] and was assumed to be complete up to 2.5 MeV. No experimental value for neutron resonance spacings ($D_0$) exists for the nucleus of interest, therefore the $\rho(S_n)$ had to be extracted from theoretical models with a resulting larger systematic uncertainty. Using the tabulated NLD from \[49\], an energy shift was applied, as described in the original publication, to match the low-lying level scheme. The energy shift upper and lower limit values were $-0.4$ and $-0.8$ MeV, respectively. The middle value used a shift of $-0.6$ MeV. Using these shifts, the upper, middle and lower values for $\rho(S_n)$ were 140, 110, and 90 MeV$^{-1}$, respectively, and these were used as limits in the normalization procedure. For consistency, the same model and energy shifts were used for extracting the $D_0$ values, 37, 39 and 41 keV, respectively. In addition, other NLD models were investigated, such as the Goriel et al. tabulated level densities \[38\].
which resulted in a $\rho(S_n)$ of 98 MeV$^{-1}$, and also the Generalized super fluid model (GSM) \cite{36,37}, which gave a $\rho(S_n)$ of 140 MeV$^{-1}$, following the procedure described above to match the low-lying discrete levels. The results of both NLD models are within the upper and lower limits mentioned above. These limits are shown in Fig. 5(a) together with the low-lying discrete levels and the experimental data. The experimental data are in good agreement with the discrete levels at low energies, however, the high energies present significant statistical fluctuations. Nevertheless, this experimental result is still useful for constraining the $\gamma$SF as described below.

![Graph showing NLD (a) and spin distribution (b)](image)

FIG. 5: (Color online) (a) Extracted NLD for $^{69}$Ni (black dots). The low-lying levels from ENSDF \cite{48} are shown in the solid black line. The upper and lower limits for the NLD used in the TALYS calculations for the extraction of the $^{68}$Ni(n,$\gamma$)$^{69}$Ni reaction cross section are represented by the dashed blue lines, with the middle value indicated by the solid blue line. (b) The $^{69}$Ni spin distribution around the neutron separation energy as calculated by \cite{48} and \cite{39}. The shaded area shows the part of the distribution that is expected to be populated in the present experiment.

The selective spin population of $\beta$ decay was taken into account in the normalization process. Starting with the $7/2^-$ ground state of $^{69}$Co, the allowed transitions will populate $5/2^-$, $7/2^-$ and $9/2^-$ states. The $\beta$-Oslo analysis considers the NLD after the emission of one $\gamma$ ray, and therefore, the populated spin distribution is expected to be $3/2 - 11/2$, of both parities. Using the theoretical spin distribution of \cite{38} and of \cite{39}, the above spin range corresponds to 60(5)% of the total, as shown in Fig. 5(b). This fraction was taken into account in the present analysis. An additional uncertainty comes from the fact that the populated $^{69}$Ni has an isomeric state at 321 keV, which $\beta$-decays with a half-life of 3.5 s \cite{48}. The presence of this state introduces an uncertainty in the experimentally extracted excitation energy. For this reason, a new upper limit for the NLD was calculated assuming the extreme where the extracted excitation energy is off by the excitation energy of the isomeric state. This assumption is an extreme upper limit for the NLD, however, since it is unknown what fraction of the excitation energy comes from the ground state, and what fraction comes from the isomeric state, the full uncertainty was included in the analysis. This new upper limit for $\rho(S_n)$ was 225 MeV$^{-1}$.

On top of the NLD normalizations, the $\gamma$SF was also normalized using available experimental data. In the traditional Oslo method, this normalization is done using the average radiative width of levels near the neutron separation energy, $<\Gamma_\gamma>$. Since in the case of $^{69}$Ni the $\Gamma_\gamma$ is not known, the normalization was done using existing experimental data in the nickel isotopic chain. These include the Coulomb dissociation of $^{68}$Ni \cite{57} and the $\gamma$SF of $^{70}$Ni as extracted previously using the $\beta$-Oslo method \cite{28}. The results are shown in Fig. 6(a). While the absolute scaling is a result of the aforementioned normalization, it can be observed that the shape of the distribution is in excellent agreement with the previous measurements. Fig. 6(b), shows the experimental middle values (stars), as well as the upper and lower limits of the $\gamma$SF, as extracted using the aforementioned upper and lower limits of the NLD. These are compared to the four $\gamma$SF models from the TALYS package that are included in Table I. It can be observed that the Kopecky-Uhl generalized Lorentzian model (strength 1) \cite{40} reproduces the shape of the experimental data very well. For this reason, this model was scaled up by a factor of 3.5 to represent the lower limit of the experimental data and by a factor of 9 to represent the upper limit of the experimental data (shaded area). It can be observed that while the other
FIG. 6: (Color online) $^{69}$Ni $\gamma$-ray strength function. (a) The central values from the present work (stars) are compared to recent experimental [28, 57] and theoretical [58] results. (b) The experimental results (central values and upper/lower limits) are compared to the theoretical models from the TALYS package, as presented in Table I. The shaded area shows the upper and lower limits of the $\gamma$SF that was used as input in the TALYS calculations.

three models presented in the figure are in fair agreement with the data above $\approx 3$ MeV, large discrepancies are observed at low energies. In addition, it should be noted that for simplicity, the default scaling of the $\gamma$SF in the TALYS code (parameter $g_{\text{norm}}$) was set to 1 for the comparison in Fig. 6(b).

Using the extracted NLD and $\gamma$SF for $^{69}$Ni, as well as the upper and lower limits, the cross section and reaction rate of the $^{68}$Ni($n,\gamma$)$^{69}$Ni were calculated. These are shown in Fig. 7(a) and (b), respectively. In both panels, the dark shaded area represents the upper/lower limits of the present work, while the middle value is shown in the solid/black line. The blue/dashed line presents theoretical calculations using the code Non-Smoker [59] (Fig. 7(a)), which also corresponds to the JINA-REACLIB results [60] (Fig. 7(b)). The dot-dashed line shows the recommended rate of the reaction library BRUSLIB [61]. The light shaded area shows the upper and lower limits of default TALYS calculations using the different models for NLD and $\gamma$SF listed in

FIG. 7: (Color online) $^{68}$Ni($n,\gamma$)$^{69}$Ni cross section (a) and reaction rate (b).
Table I. Both reaction libraries used in astrophysical calculations, JINA-REACLIB and BRUSLIB, are in good agreement with the extracted experimental reaction rates, with the JINA-REACLIB recommended rate overlapping with the experimental range, and the BRUSLIB rate being slightly lower than the lower limit.

The impact of the neutron optical model potential on the present result was also investigated. For this, the two available nOMP models in TALYS were used, namely the semi-microscopic OMP of Jeukenne-Lejeune-Mahaux (JLM) [62], and the phenomenological model of Koning and Delaroche (KD) [63]. The cross sections calculated with the two OMP models were different at most by 10%, well within the uncertainties of the present result.

The experimentally constrained reaction rate of the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction was used in astrophysical calculations for low entropy $r$-process scenarios that mainly produce the $A\approx80$ peak (weak $r$ process). Such scenarios are the accretion disk outflows that accompany a neutron star [13] or black hole-neutron star merger [64]. The weak $r$-process parametrization was done with the following wind conditions: entropy $s/k=10$, time scale = 0.1 s, and electron fraction $Y_e = 0.32$, similar to [13, 64]. The results of these weak $r$-process calculations are shown in Fig. 8. The black dots in Fig. 8(a) correspond to solar $r$-process residuals taken from [5]. The blue lines represent the calculated abundances when varying the rate of the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction within its theoretical deviations, while the red shaded area shows the impact of the experimental results. Fig. 8(b) shows the same calculations in the form of a ratio between the upper and lower limits. Both figures show that this particular rate has a local impact in the $A=69-70$ mass region and a smaller global impact, especially around $A=120$. The uncertainty coming from this reaction is significantly reduced by the present result and is now far below the observation uncertainties for mass 70 nuclei.

IV. CONCLUSIONS

The cross section and reaction rate of the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction have been experimentally constrained for the first time. The $\beta$-Oslo method was used for extracting the nuclear level density and the $\gamma$-ray strength function of $^{69}\text{Ni}$, which was populated in the $\beta$ decay of $^{69}\text{Co}$. This experiment provided the first experimental information on these two quantities for $^{69}\text{Ni}$, and they were used as input in statistical model calculations for constraining the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction. The various sources of systematic uncertainty were investigated, and the final uncertainty on the reaction rate was a factor of $\approx 3$. This is a significant reduction compared to the theoretical deviations of a factor 35. The present data reduces the uncertainties in the abundance predictions from the $^{68}\text{Ni}(n,\gamma)^{69}\text{Ni}$ reaction to less than 2.5% across the mass range.
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