Gamma production cross section measurements for $^{76}$Ge via (n,n'γ) at GELINA


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The mission of the JRC-IRMM is to promote a common and reliable European measurement system in support of EU policies.
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1. INTRODUCTION

Neutrinoless double beta decay (0νββ), the process that converts two neutrons into two protons with the emission of two electrons only, is a subject under extensive investigation, due to its importance in understanding the nature of the neutrino, exploring physics beyond the standard model [1].

Due to its very long expected half-life ($10^{25}$ y) in the promising case of $^{76}$Ge, the many experiments exploring 0νββ need to achieve a significant level of sensitivity, which involves set-ups with very large detector volumes and the simultaneous elimination of background events. Enriched $^{76}$Ge Germanium is a standard choice for detector material, since it can be produced in large sizes and offers high energy resolution and excellent efficiency.

An important setback in the use of $^{76}$Ge enriched detectors for 0νββ experiments is the existence of a γ-ray with energy 2040.7 keV originating in the 69th level of $^{76}$Ge at 3951.9 keV. This transition can create an artificial signal too close to the energy where valid 0νββ events are expected (2039.0 keV) and can easily interfere with the measurements. For this reason it is critical to have a reliable evaluation of the magnitude of this signal, which requires accurate measurements for the production cross section of the 2040.7 keV γ-ray.

One of the experiments currently investigating the neutrinoless double beta decay using enriched Ge detectors is GERDA [2], located at Gran Sasso National laboratory in Italy. The group from the Institute for Nuclear and Particle Physics of TU-Dresden participating in the GERDA project, proposed a measurement of the 2040.7 keV transition of $^{76}$Ge to take place at IRMM.

The measurement was designed to be conducted with the GAINS gamma-array for inelastic neutron scattering at GELINA, developed with the purpose of accurately measuring cross sections using the (n,n'γ) - technique [3, 4]. The experimental work was planned to be carried out in two phases:

1. an initial two-week period to observe and measure the strongest transitions of the 69th level of $^{76}$Ge using a $^{76}$Ge enriched target
2. a more extended measurement with a $^{76}$Ge–depleted target, to evaluate the effect of inelastic neutron scattering in the Ge detectors.

The first phase of the measurement campaign, described here, will be part of the PhD thesis of Alexander Domula from TU-Dresden.
2. DESCRIPTION OF THE EXPERIMENT

The present data were collected with the GAINS set-up (Gamma array for inelastic neutron scattering) [3] at the GELINA white neutron source for a total of 165 hours during April and May 2011. The set-up is located at flight path 3 of the GELINA facility, at \( \approx 200 \) m from the neutron source (198.7 m between neutron source and target).

GELINA was operated at 800 Hz repetition rate providing pulses of \(<1\) ns FWHM. The incident energy of the neutrons was determined by the time of flight technique, using the time interval between the generation of neutrons at the source and the detection of the gamma rays produced by inelastic scattering in the target.

GAINS consists of 12 large volume HPGe Canberra detectors in angles of 110\(^\circ\), 125\(^\circ\) and 150\(^\circ\) to the beam, with 4 detectors positioned at each angle at distances of 16 – 18.6 cm from the target. The GAINS detectors have typical gamma energy resolutions of \(\approx 2.3\) keV for the 1.332 MeV peak of \(^{60}\)Co. For the current measurement only the eight detectors positioned at 110\(^{\circ}\) and 150\(^{\circ}\) were used.

The normalization of the measurements is provided by a \(^{235}\)U fission chamber (FP3/200m) located upstream from the target (distance target – centre of FP3/200m: 146.8 cm), described in [3], [5].

The data acquisition involved Acquiris DC440 digitizers with 12 bit resolution and a sampling rate of 440 MS per second. The system is described in detail in [4]. Data analyzed with the GAINS setup typically have a time resolution of \(\approx 10\) ns, corresponding to the sum (9.6 ns) of four time bins of the digitizers. This results in a neutron energy resolution of about 1 keV at 1 MeV.

2.1 Samples

Two \(^{76}\)Ge enriched samples were available for the measurement:

- Sample 1 (Figures 1a, 1b), provided by TU-Dresden, was a mono-crystal Germanium slab of roughly conical shape enriched to 87.44\% (Table 1).
- Sample 2 (Fig. 1c), supplied by IRMM, had a quadrilateral shape and average thickness of 0.353 cm. The isotopic ratios stated in Table 1 are derived from combining the values of [6] and mass spectroscopy measurements of the \(\text{GeO}_2\) powder before reduction and zone refinement. The masses of the samples were measured as 14.56 g and 17.43 g respectively.

The two pieces were attached together at the target position to increase the available amount of material (Figure 1d).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(^{\text{naf}})Ge</th>
<th>(^{\text{enr}})Ge – Sample 1</th>
<th>(^{\text{enr}})Ge – Sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{70})Ge</td>
<td>21.23 ± 0.04 %</td>
<td>0.001 %</td>
<td>0%</td>
</tr>
<tr>
<td>(^{72})Ge</td>
<td>27.66 ± 0.03 %</td>
<td>0.027 %</td>
<td>0.03%</td>
</tr>
<tr>
<td>(^{73})Ge</td>
<td>7.73 ± 0.01 %</td>
<td>0.110 %</td>
<td>0.13%</td>
</tr>
<tr>
<td>(^{74})Ge</td>
<td>35.94 ± 0.02 %</td>
<td>10.350 %</td>
<td>12.3(3) %</td>
</tr>
<tr>
<td>(^{76})Ge</td>
<td>7.44 ± 0.02 %</td>
<td>87.44(6) %</td>
<td>87(1) %</td>
</tr>
</tbody>
</table>

Table 1. Isotopic abundances in natural Ge and in the enriched material of the samples.
The Sample 2 was measured on a low-background HPGe-detector before the irradiation and no gamma-emitting impurities could be detected. A day after the irradiation stopped the Sample 2 was measured again on a low-background HPGe-detector at IRMM and no activation products could be detected.

### 2.2 Gamma-detector efficiency

The determination of the gamma detection efficiency was performed by a method combining calibration measurements and Monte Carlo modeling, described in detail in refs. [5], [7]. The calibration measurements were taken with a $^{152}$Eu point source between October 2010 and February 2011 for a total of 25560(1) s. The uncertainty associated with the source was 0.8%.

The $^{152}$Eu gammas at 779, 1212 and 1408 keV were used to extrapolate to the gamma energies of interest. Taking into account the level of agreement between calculated and measured efficiencies, the overall uncertainties for the gamma detection efficiency were between 1.5-1.7%.
2.3 Neutron flux, FP3/200m efficiency

The technique for the determination of the fission chamber efficiency ([5]-[9]) is based on the rejection of the alpha peak from the amplitude spectrum with the application of a threshold in the centre of the plateau between alpha and fission fragment peaks (Figure 2). A flat or linear fit of the plateau region is then extrapolated to zero pulse height to calculate the total number of fissions. Further corrections are applied to account for the polarity effect [8] of FP3/200m, the fission fragments that stop in the deposit [10] and the inhomogeneity of the UF\textsubscript{4} foils.

For the current measurement the selected threshold applied on the amplitude spectrum resulted in an efficiency of 82(1) %.

![Figure 2. Left: schematic representation of the FP3/200m fission chamber. Right: experimental (black) pulse height distribution from FP3/200m and the calculated fission fragment spectrum (red).](image)

Due to the limited data dating time the neutron time of flight spectra had very low statistics, which resulted in many zero value channels in the neutron yield distribution. To prevent complications in the subsequent data analysis, the current spectra were normalised to the neutron flux distribution of an earlier measurement (\textsuperscript{23}Na(n,n'\gamma)) with significantly better statistics. The normalisation factor was calculated according to the fission fragment integrals above threshold for the two pulse height spectra.

2.4 Gamma production, level and total inelastic cross sections

The primary measured quantities with GAINS are differential $\gamma$-ray production cross sections, which generate total $\gamma$-production cross sections through angular integration [3]. These, in turn, give the level production cross sections using level information from the evaluated nuclear structure data files [11]. Only one observed gamma per level is used for this purpose.

Very few experimental data exist for \textsuperscript{76}Ge, and the excitation functions (up to the 39\textsuperscript{th} level) included in ENDF / B-VII.0 are based on calculations.

In the current measurement we are only interested in the 69\textsuperscript{th} level of \textsuperscript{76}Ge at 3951.89 keV and the aim is to observe and measure any of the gammas of Table 2. The level has a threshold of 4003.89 keV.
The production rate for this level is anticipated to be very low, so the focus is in the search of the strongest transitions of 3951.7, 3388.75 and 2843.5 keV, and possible escape peaks from these gammas.

<table>
<thead>
<tr>
<th>Eγ (keV)</th>
<th>Iγ</th>
</tr>
</thead>
<tbody>
<tr>
<td>1259.9(5)</td>
<td>7(2)</td>
</tr>
<tr>
<td>2040.70(25)</td>
<td>8(2)</td>
</tr>
<tr>
<td>2843.5(9)</td>
<td>38(2)</td>
</tr>
<tr>
<td>3388.75(12)</td>
<td>67(4)</td>
</tr>
<tr>
<td>3951.70(14)</td>
<td>100(8)</td>
</tr>
</tbody>
</table>

Table 2: Gammas associated with the 69th level of 76Ge [11].

The acquired pulse height spectra showed gamma peaks of all Ge isotopes in the target material, with well defined lines for 76Ge at least up to 23rd excited state. However, none of the transitions of Table 2 were clearly observable (Figure 3). Because of the poor statistics of the measurement, a more thorough analysis was deemed necessary to investigate whether any of these gammas are actually present in the spectra.

For this reason a detailed calibration was made for all detector spectra, using past 152Eu measurements and 10 background and 76Ge gammas with energies ranging between 431 and 2920 keV. The full width at half maximum of the peaks was also taken into account. Based on this calibration, upper and lower limits were calculated for the gammas of Table 2 (Figure 3) and for the regions to be used for background subtraction.

The yields and production cross sections were then calculated for the 1259.9, 2040.7, 3388.75 and 3951.7 keV gammas of the level, according to the standard procedure described in [3].

Two more 76Ge gammas were investigated as a reference (Table 2), the 562.93 keV gamma from the 1st excited state to the ground state and the 545.51 keV transition from the 2nd to the 1st excited state. As an additional check, the yield of the 1460.8 keV line of 40K was also examined, and confirmed to be constant and independent of neutron energy.

Table 3 shows a summary of the studied 76Ge(n, n'γ) gammas and associated levels. The contributions of the observed gammas to each level are related to the weights displayed in Table 4, according to the expressions of reference [3]. One observed transition is used for the calculation of each level cross section, so the 3951.89 keV level can be produced separately from each of the four related gammas.

<table>
<thead>
<tr>
<th>Eγ (keV)</th>
<th>Elevel1 (keV)</th>
<th>Jν1</th>
<th>T1/2</th>
<th>Elevel2 (keV)</th>
<th>Jν2</th>
<th>Iγ</th>
<th>γ mult.</th>
</tr>
</thead>
<tbody>
<tr>
<td>562.93(3)</td>
<td>562.93(3)</td>
<td>2+</td>
<td>18.2(2) ps</td>
<td>0</td>
<td>0+</td>
<td>100</td>
<td>E2</td>
</tr>
<tr>
<td>545.51 (3)</td>
<td>1108.44(4)</td>
<td>2+</td>
<td>8.0(15) ps</td>
<td>562.93(3)</td>
<td>2+</td>
<td>100.00(2)</td>
<td>E2+M1</td>
</tr>
<tr>
<td>1259.9(5)</td>
<td>3951.89(7)</td>
<td>(1,2+)</td>
<td>2692.40(8)</td>
<td>3-</td>
<td>7(2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2040.70(25)</td>
<td>3951.89(7)</td>
<td>(1,2+)</td>
<td>1911.07(11)</td>
<td>0+</td>
<td>8(2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3388.75(12)</td>
<td>3951.89(7)</td>
<td>(1,2+)</td>
<td>562.93(3)</td>
<td>2+</td>
<td>67(4)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3951.70(14)</td>
<td>3951.89(7)</td>
<td>(1,2+)</td>
<td>0.0</td>
<td>0+</td>
<td>100(8)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3: Examined gammas from 76Ge(n,n'γ) and associated initial (1) and final (2) levels [11].
Table 4: Contributing weights of the observed gamma production cross sections to the construction of the level cross sections (See reference [3] for a description of the use of these weights).

<table>
<thead>
<tr>
<th>γ-ray\Level</th>
<th>562.93</th>
<th>1108.44</th>
<th>3951.89</th>
<th>3951.89</th>
<th>3951.89</th>
<th>3951.89</th>
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<tr>
<td>562.93</td>
<td>1.000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>545.51</td>
<td>-1.00(6)</td>
<td>1.680(71)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1259.9</td>
<td>-10(3)</td>
<td>-5(2)</td>
<td>31(9)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2040.70</td>
<td>-8(2)</td>
<td>-5(1)</td>
<td>28(7)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3388.75</td>
<td>-1.00(81)</td>
<td>-0.567(47)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3951.70</td>
<td>-0.670(51)</td>
<td>-0.38(3)</td>
<td>2.20(11)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 3. The software gates applied on the amplitude spectra of one of the GAINS detectors to produce the gamma yields of the studied transitions.
3. PRESENTATION OF RESULTS

The results presented here are produced from the full dataset and are corrected for multiple scattering and attenuation effects. These corrections were calculated with the help of iterative MCNP5 simulations, starting with the \( \gamma \)-production cross sections for \(^{76}\text{Ge}\) obtained from the current measurement. Good convergence was obtained with two iterations.

Figure 4 shows the angle-integrated gamma production cross-sections for the transitions 562.93 and 545.51 keV of the first two excited states and the resulting level cross sections. For both gammas the region below threshold is dominated by noise, and there is a clear rise at the level's threshold. For the 562.93 keV transition the cross section reaches 1 b at about 1 MeV and slightly exceeds it above that energy. The 545.51 keV gamma does not go much above 400 mb. Due to the low statistics of the measurement, both cross sections involve high uncertainties, averaging 12% and 16% respectively for the greatest part of the examined energy region.

The calculated level cross section of the first excited state is absolute below the threshold of the second excited state, at 1123 keV. Up to that energy there is reasonably good agreement with existing data [12, 13], in spite of the large uncertainties (average 17%) of the current measurement.

Figure 4. The measured gamma production cross sections for the 562.93 and 545.51 keV transitions (top) and the related level cross sections (bottom) of the first two excited states of \(^{76}\text{Ge}\). For the 562.93 keV level existing data from [12] and [13] are displayed for comparison.
The above confirm that the analysis procedure and parameters are correct and acceptable results can be achieved even with the current statistics.

![Graphs showing gamma production cross sections for three transitions of the 3951.89 keV level.](a)

![Graphs showing gamma production cross sections for three transitions of the 3951.89 keV level.](b)

![Graphs showing gamma production cross sections for three transitions of the 3951.89 keV level.](c)

**Figure 5.** Measured gamma production cross sections for three of the transitions of the 3951.89 keV level.
For the gammas associated with the 3951.89 keV level, the situation is no longer clear, as shown in Figure 5. The distributions have been plotted using uniform bin width on either side of the threshold (2-5 MeV) in order to evaluate possible variations, and much broader bins in the high energies for a better assessment of general trends.

In the case of the strongest 3951.70 keV transition (Fig. 5a) the cross section appears to rise above zero at the level threshold of 4003.9 keV. Beyond that value it seems to have a positive bias, however negative values appear in the high energies even with very coarse binning.

For the 3388.75 keV gamma (Fig. 5b) a shift appears to begin at threshold but towards negative values, and for the 2040.70 keV transition (Fig. 5c) a similar but stronger dip begins at about 3 MeV and seems to reach a minimum at threshold. Above threshold none of these gammas show any significant positive trend, and overall these distributions are consistent with zero.

Because of the way that these invisible peaks were gated (Fig. 3), it would be very useful to make some additional tests by shifting the peak and background limits by a few channels on either direction and repeating the analysis. A few such checks would give a better idea on whether the limits were defined correctly, if there is a definite change at threshold and measurable cross sections above it. In the case that the zero result is verified, upper limits can be defined for the cross sections.

3.1 The data and how to use them

The delivered data files contain five of the gamma production cross sections of Table 2, with the designation nnnnGpxs1.his, where nnnn the gamma-ray energy.

Each .his file has four columns. The first is the time-of-flight in ns, the second the energy in keV, the third the cross section in b and the fourth the uncertainty of the cross section in b.

The required software for the continuation and/or extension of the analysis was also supplied, together with detailed instructions on how to use it and a step-by-step description of the full analysis process (file Analysis_info.doc), starting with the raw data. All necessary input files are provided, as well as a few examples of output and batch files.
4. SUMMARY AND OUTLOOK

The inelastic scattering data obtained at IRMM with the GAINS setup at the GELINA time-of flight facility were delivered to the Institute for Nuclear and Particle Physics of TU-Dresden in the form of data files containing gamma production cross sections. The investigated $\gamma$-rays, originating in the 69$^{th}$ excited state of $^{76}$Ge, are of interest in the research on neutrinoless double beta decay.

The results do not show sufficient evidence of measurable cross sections for the studied transitions. The distribution of the strongest gamma at 3951.70 keV shows the expected increase at threshold and a positive bias, however negative values exist at high energies. The two weaker transitions at 3388.75 and 2040.70 keV show no discernible positive trend and questionable shift at threshold.

Overall we cannot confirm that the population of the 3951.89 keV level was observed in the current measurement.

As an attempt to draw some quantitative conclusions from the present results, the cross section of the 3951.70 keV gamma does not go over $\sim 40$ mb (within $1\sigma$) for all energies above threshold. According to Table 2 that gives an upper limit of 3.2 mb for the 2040.70 keV $\gamma$-ray. Similarly, we can estimate a cross section $\leq 25$ mb for the 3388.75 keV gamma, which gives $\leq 3.0$ mb for the 2040.70 keV transition.

To effectively measure cross sections of this scale much better statistics are required, and a measurement at a Van de Graaf with high intensity monoenergetic neutron beams would be highly advantageous. It is also recommended that further analysis of the current data is carried out with a more detailed examination of the data analysis parameters.
REFERENCES

Abstract
This report describes the inelastic scattering data delivered to the Institute for Nuclear and Particle Physics of TU-Dresden. The GAINS setup was used for measurements of gamma production cross sections associated with the 69th excited state of $^{76}$Ge, using the $(n, n'\gamma)$ technique. The experimental work was performed at the GELINA facility at a 200 m flight path with eight high purity germanium detectors, using highly enriched $^{76}$Ge samples. A brief description of the experimental details and the results are presented.
The mission of the JRC is to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of EU policies. As a service of the European Commission, the JRC functions as a reference centre of science and technology for the Union. Close to the policy-making process, it serves the common interest of the Member States, while being independent of special interests, whether private or national.