Targets of ^{72}Germanium for in-beam gamma-ray spectroscopy

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Abstract. At the Experimental Storage Ring ESR at GSI experiments on the investigation of the isolated nuclear two-photon decay were performed [1]. For a better understanding, complementary experiments were planned at the TANDEM accelerator at the University of Cologne, Germany. For these experiments targets from enriched ^{72}Ge on a thin titanium backing as well as self-supporting targets were needed. As we had only scarce material available, thermal evaporation in a close evaporation geometry was the method of choice. ^{72}Ge-targets in a thickness range of 350 µg/cm² - 370 µg/cm², self-supporting and with titanium backing, were obtained. We report on the target production and on first results of their application in the experiment.

1 Introduction

In 2023, storage-ring experiments to investigate the nuclear two-photon (2γ) decay on ^{72}Ge-isomers were performed with very high precision. These experiments revealed a partial half-life for the 2γ decay of the 0+ isomer that is a factor of ~ 10 shorter than was expected from the extrapolation of previous γ-spectroscopy experiments on ^{16}O, ^{40}Ca, and ^{90}Zn [1-3].

Since with the storage-ring experiments only the life-time of the isomer could be measured, future direct γ-spectroscopy experiments in the low-energy regime would be favourable. Due to the now expected larger branching ratio of the 2γ decay channel such experiments should be possible with state-of-art γ-arrays.

In 2023, the opportunity arose at short notice to conduct a complementary experiment to study the nuclear two-photon decay with proton scattering on ^{72}Ge-targets at TANDEM at the University of Cologne. The aim here was to estimate from the measured γ-background the possibilities and requirements for future experiments with direct γ-decay studies.

2 Requirements and prerequisites

For the proton-scattering experiment at the TANDEM experiment ^{72}Ge-targets with a thickness of about 500 µg/cm² were requested. The tantalum frames had outer dimensions of 10 mm x 25 mm and an effective target area of 0.91 cm². For this thickness the energy loss of the backscattered protons is minimum which means that the resolution is maintained. For thicker foils the reaction rate is higher, but the energy resolution gets poorer.

In order to separate the comparable small peaks of the 2γ decay from the other reactions taking place, all additional sources that could contribute to the γ-background had to be minimized. This led to the following measures and requirements:

- The target frames were made of tantalum, which is known to only have a low γ-excitation in case the frame is hit in the experiment accidentally.
- The target material had to be as pure and as highly enriched as possible to avoid background from the contamination or other germanium isotopes.
- The targets should preferably be self-supporting. However, since the foils prepared with a water-soluble interlayer are in contact with water during the floating procedure, oxygen-contamination could become a problem. Non-water-soluble parting agents were not taken into consideration because this would have required longer time for process development.
- Therefore, also ^{72}Ge-targets on a thin titanium backing were required as a backup.

To be on the safe side, two to three targets with and without backing were requested that had to be produced within 4 weeks, including the rolling of titanium backings of about 2 µm in thickness.

3 Preliminary studies

3.1 Properties of germanium

Germanium is a lustrous, hard-brittle metalloid, which is chemically similar to silicon and tin. It is a semiconductor with covalent bonds. The melting temperature is 938 °C and the density 5.327 g/cm³. Germanium has four stable isotopes and the here relevant isotope ^{72}Ge has a natural abundance of 27.4 %. [4].

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3.2 Published deposition methods

Maxman [5] prepared thin germanium targets by thermal evaporation from a tantalum boat.

From the paper of Barna et al. it is known that thick germanium deposits tend to crack and peel [6]. Different techniques are used for preparation depending on the application and the capabilities of the respective laboratories; Méens and Ehret [7] published a review on the methods used for germanium deposition. However, there are only few reports on deposition of enriched material and almost no information on dimension and homogeneity of the deposits or yields of the process.

4 Tests and decisions

With this knowledge, we started test runs with natural germanium. We used a standard setup for thermal evaporation from a tantalum boat. We deposited the germanium on glass substrates that were covered with betaine-sucrose as parting agent and on 2 µm thick titanium backing, as will be described in section 5.

We experienced that germanium layers thicker than about 500 µg/cm² tend to sink under water while separating from the substrate, which makes is difficult or even impossible to mount them on a frame without damaging. Therefore, we aimed for a thickness 400 – 500 µg/cm² for the enriched targets.

We only had 300 mg elemental ^{72}Ge with a high enrichment of 98.2 % on stock. The upcoming beam determined our production period, so purchasing more starting material was no option.

5 Target production

In order to make do with the limited amount of enriched germanium, we decided to mount the betaine-sucrose-coated glass substrate with dimensions of 55 mm x 55 mm in the centre of the sample holder and additionally attached two frames with titanium backing sideways on the same holder. In this way we could coat the Ti-backed frames and the coated glass substrate in one run, as shown in figure 1.

Fig. 1. Sample holder with the mounted glass substrate, already coated with betaine-sucrose in the middle and two target frames with titanium backing attached sideways.

The substrate holder is mounted on a rotation drive in a distance of about 200 mm above the tantalum boat, as depicted in figure 2. The rather large distance is necessary to prevent the betaine-sucrose layer from temperature damage, however the yield is significantly reduced. The sample holder is rotated during evaporation for a better homogeneity.

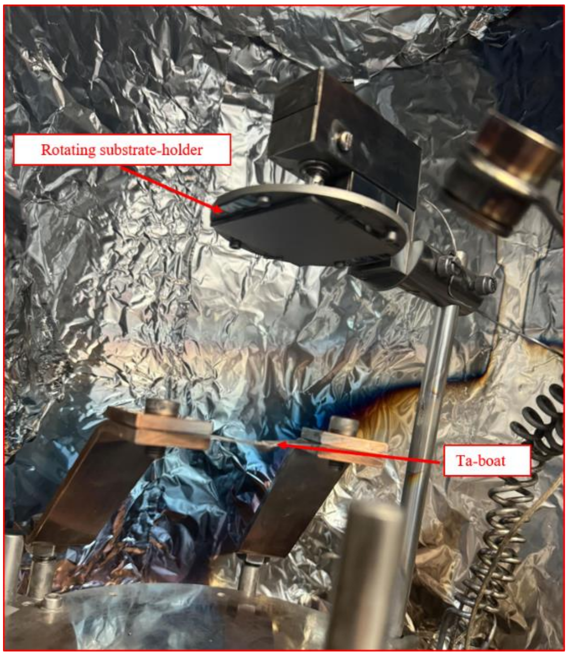

Fig. 2. Vacuum chamber with tantalum boat and mounted substrate holder on a rotation drive.

The thickness of the target layer is determined by the weighted sample of the target material; the evaporation process ends when the target material is totally evaporated from the boat. The amount of starting material that is necessary for the wanted layer thickness was estimated in preceding test runs with natural germanium.

For these tests a coated glass substrate was mounted in the evaporator and the total amount of starting material was evaporated from the tantalum boat. The target layer was then scratched to pieces appropriate for the target frames and floated in distilled water. Then the foils were fetched on the tantalum frames.

The empty frames had been weighed in advance with a micro balance and are then weighed again with the target foils attached. The difference in weight together with the known area of the frame gives the thickness of the target foil. In the following run the initial weight of the crucible was corrected accordingly.

With this method we step by step estimated an amount of necessary starting material of 100 – 150 mg for a target thickness of about 400 µg/cm². However, it is important to note that the natural and the enriched material do not necessarily evaporate in the same way.

While we could theoretically get 8 targets from one substrate plate, practically only 3 - 4 could successfully be mounted on frames, because of brittleness or sinking of the floated foils.

We conducted two evaporation runs with ^{72}Ge. In the first production run we had a weighted sample of 100 mg ^{72}Ge and increased the weighted sample to 130 mg for the second production run.

The results are summarized in table 1. As can be seen by the values, the target thickness achieved with enriched material tends to be smaller than estimated by the tests with natural germanium. Only taking into account the active target area of 0.91 cm² a total yield of the process between 1 and 2 % is achieved, depending if we count only the self-supporting targets or also the ones with titanium backing.

Table 1. Number and thickness of targets delivered.

Weighted sample	Target thickness	Number of targets	
		Self-supported	**On Ti-backing**
100 mg	**350 ± 20 µg/cm²**	**3**	**2**
130 mg	**370± 20 µg/cm²**	**4**	**2**

A photo of two self-supporting ^{72}Ge-targets glued to the frames is depicted in figure 3.

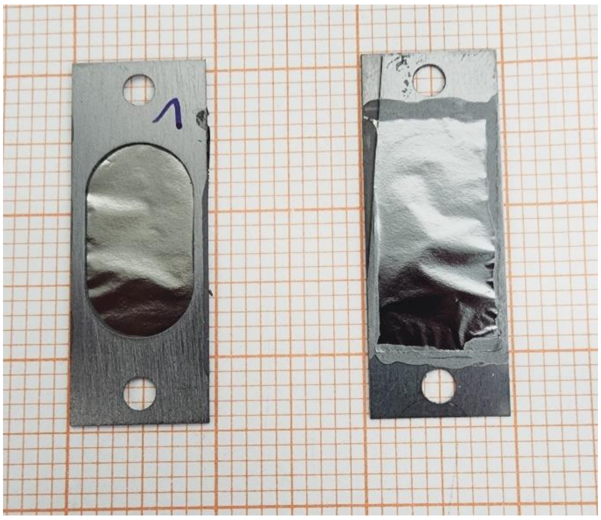

Fig. 3. Self-supported targets of 72Ge-targets on HORUS-frames with a thickness of ~ 350 µ/cm².

6 Application

The ^{72}Ge-targets were sent to Cologne and were applied in the proton scattering experiment. In figure 4, the target ladder of the experiment is shown with one of ^{72}Ge-targets mounted in the middle position. Left of it a gold target is mounted for calibration of the particle detectors and right of it an aperture for focussing the beam.

The experiment aimed to study the atomic 2γ decay with proton scattering and to measure the γ-background. In figure 5, one of the measured proton spectra with beam energy of 5 MeV is shown. On the far right of the spectrum at 5000 keV you see the large peak of the elastically scattered protons. On the left, you see two well-separated peaks from the inelastic scattering showing the first two excited states of the ^{72}Ge, proving the population of the state in question. The small peak to the very left shows (atomic) electrons, which are emitted in the de-excitation of the state of interest. From the intensity of observed peaks, it can be estimated what gamma efficiency would be necessary for a successful future γ-spectroscopy experiment.

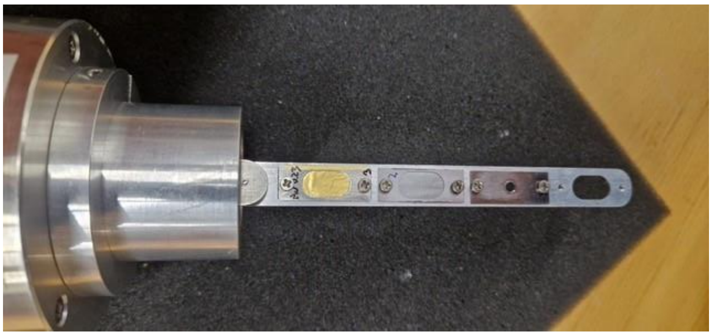

Fig. 4. Target ladder of the TANDEM experiment:

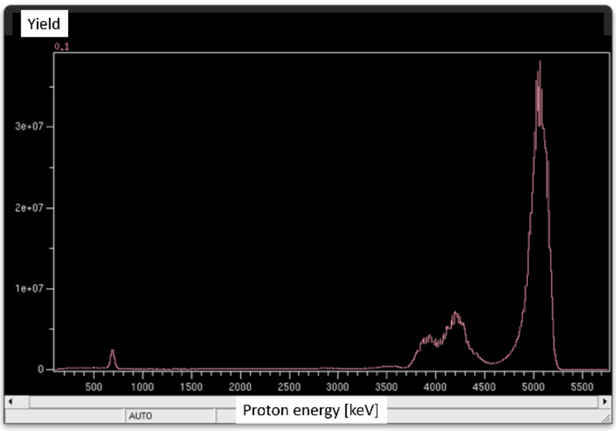

Fig. 5. Proton spectrum from the irradiation of the ^{72}Ge target with a beam energy of 5 MeV.

7 Summary

We successfully produced ^{72}Ge-targets by thermal evaporation with a thickness of ~ 360 µg/cm² in a very short time frame and with a limited amount of enriched starting material available. We delivered self-supporting targets as well as targets on titanium backing as a backup, well in time. Due to the tight time schedule the process was not optimum and has to be improved for future productions.

The ^{72}Ge-targets were successfully applied in a proton scattering experiment at the University of Cologne and enabled an estimation for the γ-detector efficiency necessary for future investigations on the nuclear 2γ decay.

The targets on titanium backing were not applied in the experiment since the purity of the self-supported targets proved to be suitable for the experiment.

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