Application of Calorimetric Low-Temperature Detectors for the Investigation of Z-Yield Distributions of Fission Fragments

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Abstract In recent experiments, the new concept of calorimetric low-temperature detectors (CLTDs) was applied for the first time for the investigation of isotopic yields of fission fragments. Fragments from neutron-induced fission were mass-separated by the LOHENGRIN spectrometer at the ILL Grenoble and, after passing silicon nitride membranes used as degraders, detected in a CLTD array. The concept of a CLTD provides a fundamental advantage over conventional ionization-mediated detectors, in particular for heavier particle masses at low energies. Using fissile targets of 235U, 239Pu and 241Pu, nuclear charge separation was studied for selected masses in the region 82 ≤ A ≤ 139. For light fragments, the Z-resolution matches historically best values with conventional techniques, while for heavier masses substantial improvement was attained. We have gained first LOHENGRIN data on the isotopic yields in the range A = 91 to 112 of 241Pu(n,f). Towards mass-symmetry, known Z-yield data were extended to the range A = 110 to 113 for 239Pu(n,f). Extended data sets were cumulated for A = 92 and 96 because of a recent request from studies on the reactor antineutrino spectrum. Furthermore, considerable progress was achieved to extend isotopic yield measurements up to the heavy-mass region, hardly accessible until now.
Keywords Bolometer for heavy ions · Thermal neutron-induced fission · Z-yield distributions

1 Introduction

Precise data on the characteristics of fission-fragment yield distributions in terms of mass, nuclear charge, and kinetic energy are of great interest, on the one hand, for a better understanding of the fission process and, on the other hand, in applied fields, e.g. for calculating accumulation and inventory of fission products at various stages of the nuclear fuel cycle in a reactor [1, 2]. Since more than four decades, the recoil mass spectrometer LOHENGRIN [3], at the ILL Grenoble, has been a leading instrument for fission-fragment studies. Fission fragments emerging from a thin fissile target located close to the high-flux reactor core (i.e. at a thermal neutron flux $\approx 5 \times 10^{14}$ n cm$^{-2}$ s$^{-1}$) are separated according to the chosen ratios $E/q$ and $A/q$ ($E =$ kinetic energy, $A =$ fragment mass, $q =$ ionic charge) and detected with suitable energy detectors (usually silicon detectors or ionization chambers). For determining isotopic fragment yields, a fairly universal method is the passive absorber technique exploiting the Z-dependent energy loss of fission fragments in an energy degrader [4]. Whereas this method was applied successfully in many experiments for the light-mass region [5], data are scarce in the heavy-mass region. Limitations of the detection techniques such as energy resolution and the influence of the pulse height defect of ionization detectors or the quality of the absorber material with respect to homogeneity and energy loss straggling make the determination of nuclear charge distributions in this region quite challenging or even impossible. Due to their good energy resolution and energy linearity, the use of calorimetric low-temperature detectors (CLTD’s) [6–8] promises, in combination with an adequate absorber material, an improvement for this kind of measurements.

2 Experimental Set-Up

The detector array used in the present experiment has an active area of $15 \times 15$ mm$^2$ and consists of 25 independent detector pixels with transition edge sensors (TES) operated at $T \approx 1.5$ K [6–8]. The individual pixels consist of 430-μm-thick sapphire absorbers on which Al-strip thermometers are evaporated by photolithographic techniques. Each pixel is individually temperature regulated via a gold layer evaporated on the pixel. For the readout, conventional pulse electronics consisting of low-noise preamplifiers and flash ADCs are used. The granular structure of the detector was chosen to keep the heat capacity sufficiently small in order to provide high sensitivity. The performance of the individual pixels and of the array turned out to be sufficient for successful application in the present experiment. In detail, we obtained a pulse rise time of $\sim 10$ μs, a pulse decay time of $\sim 5$ ms (allowing a maximum rate per pixel of $\sim 100$ events/s), a dynamic energy range of $\sim 10$ to 200 MeV and a noise level of $\sim 250$ keV. The energy resolution and energy linearity, which could not be obtained in the present experiment due to a degrader foil permanently mounted in front of the array, are expected to be similar as observed in previous measurements [6–8].
The $^4$He-bath cryostat containing the CLTD array was coupled, without any entrance foil, to the straight exit flange of LOHENGRIN inclined by 35° (see Fig. 1). In a first attempt [9], SiN absorbers were mounted on a movable manipulator at a distance of 95 cm outside the cryostat. This resulted in largely reduced counting efficiency due to small-angle scattering and increased the background by contaminating mass lines. To counter these problems, the experimental set-up was upgraded by installing a remotely controlled sample changer for SiN absorber foils inside the cryostat, at a distance of a few millimetres to the CLTDs.

The design of the new system is displayed in Fig. 1. The rotatable disc, with six positions for SiN foils of $16 \times 10$ mm$^2$ area each, is operated by a remote-controlled piezo-driven rotary stepper positioner. We use the system ANR240/RES from Attocube [10], which consists of a positioner with resistive encoder along with the controller ANC350. The device operates under vacuum and at temperatures as low as 10 mK and allows a reproducible positioning with an accuracy of 0.050° ($\approx 30 \mu$m for the current design). The SiN foil stacks mounted on this disc were 1, 4, 5, 6, 7 μm thick.

3 Experimental Results and Discussion

Using fissile targets of $^{235}$U, $^{239}$Pu and $^{241}$Pu, the quality of nuclear charge separation was studied for selected masses in the region $82 \leq A \leq 139$ as a function of degrader thickness and fission-fragment kinetic energies. For the light fragment group, good Z-resolution was obtained (see, Fig. 2b), sufficiently high to clearly separate individual nuclear charges in the residual-energy spectra and, as expected, with the resolving power improving towards higher energy losses. We could already match the historically best Z-resolutions, e.g. $Z/\Delta Z = 55$ at $Z = 37$ ($\Delta Z$ is defined as the ratio of the peak width in FWHM and the difference between a central Z peak and adjacent peaks in the rest-energy spectra [4]) (see, Fig. 2a), achieved conventionally with parylene-C absorbers and ionization chambers [11, 12].

Towards mass-symmetry, new LOHENGRIN data were obtained in the mass range $A = 90$ to 109 for $^{241}$Pu and known Z-yield data [13] were supplemented in the range $A = 110$ to 112 for $^{241}$Pu, and $A = 111$ to 113 for $^{239}$Pu. The investigation of isotopic yields for the heavier masses $A \geq 108$ (Fig. 2c) was started with the aim to study the onset of even−odd effects in the transition region from the light fragment group.
Fig. 2 a $Z$ resolving power ($Z/\Delta Z$) versus nuclear charge obtained with CLTDs and SiN absorbers for fission fragments at ILL and for heavy ions at MLL Garching as compared to data measured by Quade et al. [11] and Bocquet et al. (partly extrapolations) [12]. b–d Sample residual-energy spectra (preliminary) for fragments in light-, symmetry and heavy-mass regions, respectively, where $A$ is the mass, $q$ is the ionic charge and $E_{\text{in}}$ is the incident energy from LOHENGRIN (Color figure online)

Fig. 3 Fractional $Z$-yields (preliminary) for $A = 90$ to 112 for $^{241}\text{Pu}(n_{\text{th}},f)$ (Color figure online)

towards the symmetry region, which is of high interest for verification of nuclear fission models [14]. As an example, preliminary data obtained for $^{241}\text{Pu}$ are displayed in Fig. 3. A pronounced even–odd $Z$ staggering effect is observed.
For heavy fragments with $A > 128$, the obtained Z-resolution was insufficient to resolve individual peaks in the residual-energy spectra. Instead, the individual intensities have to be retrieved from constrained fitting of the overlapping peaks (see Fig. 2d), a method well established in high-precision mass spectrometry [15]. In our case, the fitting quality benefits from the outstanding properties of the CLTDs for the low-energy fission fragments after energy degradation (see above).

Preceding test measurements at the tandem accelerator at the MLL Garching with stable $^{130}$Te and $^{127}$I ion beams, aiming at determining the energy loss difference for adjacent Z values with our set-up, provided also valuable data on the peak shapes in this mass region. Here, residual-energy peaks reveal asymmetric shapes at larger degrader thicknesses, attributed to the energy loss processes. With a precise knowledge on the response function, constrained fits for the deconvolution of measured residual-energy spectra could be reliably performed, yielding a resolving power of $Z/\Delta Z = 28$ for $Z = 52$. This demonstrates a significant improvement as compared to extrapolations from previous measurements [12] (see Fig. 2a). For the $^{239}$Pu target, we can also build on recent isotopic yield measurements by $\gamma$-ray spectrometry [16]. Besides the possibility of cross-checking available Z-yield data for $A = 133, 134$ and $136, 137$ [18] with an alternative technique, we are now able to complete the experimental data sets with masses $128–132$ and $135$, which are either not easily or not at all accessible with $\gamma$-ray spectrometry.

For a precise determination of isotopic yields with LOHENGRIN, it is mandatory to measure nuclear charge distributions for different ionic charge states $q$ and kinetic energies $E$, and follow the target burn-up during the measuring period. Due to its complexity, data analysis is still in progress, and all data shown at present are considered as preliminary. Extended measurements were performed for masses $A = 92$ and $96$, for several ionic charges and 5 different energies. The isotopic fission yields of $^{92}$Rb and $^{96}$Y from $^{235}$U(n$_{th}$, f), $^{239}$Pu(n$_{th}$, f) and $^{241}$Pu(n$_{th}$, f) are of particular interest, since more precise data than available [17] have been requested recently for achieving a better understanding of the reactor antineutrino spectrum [18–20]. The decay of these isotopes is a main contributor to the integral antineutrino spectra above 4 MeV.

4 Summary and Outlook

The passive absorber method, for the first time using SiN membranes as degraders and CLTDs as residual-energy detectors, was successfully applied for determining Z-yields of fission fragments. A systematic study on Z-resolution with variable degrader thickness was performed in a wide range of fragment masses $A$ and energies $E$. For the light fragment group, the Z-resolution attained matches historically best values achieved with parylene-C absorbers and ionization chambers, while for the symmetry and the heavy-mass regions a substantial improvement was observed with the new set-up. We have gained first LOHENGRIN data on the isotopic yields in the light-mass group, $90 \leq A \leq 112$ for $^{241}$Pu(n,f). Towards mass-symmetry, known Z-yield data were supplemented in the range $A = 111$ to $113$ for $^{239}$Pu(n,f), to study odd–even staggering. Extended data sets were cumulated for the masses $A = 92$ and $96$ due to special interests in the precise yields of these isotopes for studies on the reactor
anti-neutrino spectrum [18–20]. Furthermore, considerable progress was achieved to extend Z-yield measurements to the heavy-mass region, $128 \leq A \leq 139$ which was hardly accessible until now.

We finally believe that our approach of deducing isotopic fission-fragment yields with applying the novel technologies of CLTDs and SiN degraders provides a wide scope for further improvements both from a methodical and technological point of view.

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