# APPLICATION OF CALORIMETRIC LOW-TEMPERATURE DETECTORS FOR INVESTIGATION OF Z-DISTRIBUTIONS OF FISSION FRAGMENTS

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## Abstract:

The investigation of mass, charge and energy distributions of fragments from thermal neutron induced fission is of high interest for our understanding of the fission process. In a recent experiment, performed at the research reactor at the ILL Grenoble, the new concept of calorimetric low-temperature detectors (CLTD`s) was applied for the first time for the investigation of Z-yield distributions of fission fragments. Fission fragments from $^{235}U(n_{th},f)$, were mass-separated by the LOHENGRIN spectrometer and, after passing stacks of silicon nitride ($Si_3N_4$) membranes as a novel type of degrader material, detected in an array of CLTD`s. In the present contribution, the operation principle of a CLTD will be briefly introduced, and the design of dedicated detectors for heavy ions and their performance discussed. The advantages of CLTD detectors over conventional ionization detectors (solid-state Si or gaseous detectors) are the more complete energy detection, the smaller energy gap of the detected quanta (phonons) leading to lower statistical limitation, and the absence of dead layers and entrance windows. Therefore these detectors have been proven to provide high energy resolution, good

energy linearity, low detection thresholds and insensitivity against radiation damage, a promising performance for detecting fission fragments.

The quality of nuclear charge separation was studied for selected masses in the region $82 \leq A \leq 132$ as a function of degrader thickness and fission-fragment kinetic energies. For the light fragment group (Z<43, A<107), the Z-resolution attained could already match historically best values achieved conventionally with Parylene-C absorbers and ionization chambers. Extended measurements were performed for mass A = 92, for various ionic charges and fragment energies. Here, the isotopic yield of $^{92}$Rb was of particular interest, due to a recent request from studies on the reactor antineutrino spectrum. An attempt was made to extend isotopic yield measurements to the symmetric and heavy-mass regions, hardly accessible until now. Finally a recent upgrade of the experimental set-up by installing a remotely controlled sample changer for the $Si_3N_4$ absorber foils inside the cryostat is briefly described.

## 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 for verifying fission models, and, on the other hand, required at several stages of the nuclear fuel cycle [1,2]. Since more than 3 decades, the recoil mass spectrometer LOHENGRIN [3], available 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 (thermal neutron flux $\approx 5\times10^{14}$ n cm$^{-2}$ s$^{-1}$) are separated with a certain ratio of E/q and A/q (A = mass of fragment, q = ionic charge) and registered with suitable energy detectors (silicon SB 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]. Due to its perfect mass resolution, LOHENGRIN has contributed more complete data sets on mass, nuclear-charge and energy distributions than any other method, with the isotopic yields, however, being restricted to the region of light fragment masses [5]. Instead, isotopic yields of specific radioactive isotopes in the heavy mass group were determined by high-resolution Gamma spectroscopy in combination with LOHENGRIN [6] or, formerly, by radiochemical methods [7].

In the present work we have applied a new experimental approach to determine isotopic yields at LOHENGRIN by the passive absorber method using an array of calorimetric low-temperature detectors (CLTD's) instead of an ionization chamber for the residual energy measurement. The advantages of CLTD detectors over conventional ionization detectors (sold-state Si or gaseous detectors) are the more complete energy detection, the smaller energy gap of the detected quanta (phonons), and the absence of any entrance window or dead layer, leading to substantial improvements in basic detector properties, as for example energy resolution, energy linearity, detection threshold, etc. [8 - 10].

Furthermore, stacks of commercially available silicon nitride ($Si_3N_4$) membranes [11] have been tested as degrader material, replacing the previously used Parylene-C plastic foils. Silicon nitride is a modern highly resistant material out of which stable and homogeneous membranes thinner than 100 nm can be fabricated by lithographic techniques. These membranes are extensively used outside nuclear physics, e.g., as substrates in transmission electron microscopy or ionization-chamber entrance windows for low-energy ERDA (elastic recoils detection analysis) applications [12]. We have applied, for the first time, stacks of up to seven 1000 nm thick $Si_3N_4$ membranes for nuclear-charge determination by energy degrada-

tion of fission fragments. Preceding test measurements at the tandem accelerator at the MLL Garching with stable $^{109}$Ag and $^{127}$I ion beams have shown promising results concerning Z-resolving power, with improved performance as compared to Parylene-C [13]. This is attributed to their good homogeneity, although the contribution of energy-loss straggling to Z-resolution might be slightly larger due to the somewhat heavier average atomic number.

## 2. Properties of CLTD's for particle detection

The basic principle of a CLTD is schematically displayed in fig. 1. Incident particles are stopped in an absorber located in an evacuated volume inside a cryostat. The temperature rise of the absorber due to the thermalization of the particle's kinetic energy is measured by a thermometer. Finally, on a time scale of tenths of μs, the deposited heat is fed back to a heat sink through thermal coupling. The sensitivity of such detectors is inversely proportional to their heat capacity C. Therefore, detector pixels are small in size and operated at low temperatures ($C \sim T^3$). As thermometers, thermistors with a strong temperature dependence of their resistance at their low working temperatures, for example superconducting transition-edge thermometers, are used. Since CLTD's are windowless towards the beam and signal generation is not based on ionization alone, thus leading to a more complete energy detection, they do not experience any pulse-height defect even at high energy loss density. The performance of detector pixels with a layout similar to the present one was systematically investigated in several measurements with various heavy ion beams ($^{13}$C…$^{238}$U), at incident energies ranging from 0.1 MeV/u up to 360 MeV/u [8]. For a wide range of ion species, output signal amplitudes are perfectly linear with energy, and no pulse-height defect is observed (see fig 2). Note that even at a quite low energy of 20 MeV a heavy nucleus such as $^{238}$U generates the same signal amplitude as the light $^{13}$C. At the same time, due to the good quantum statistics of phonon formation, and due to the more complete energy detection, the energy resolution is down to $\Delta E/E = 2 - 3 \times 10^{-3}$ (FWHM) for heavy ions, being not dominated by statistical effects but structural stability due to design limitations. As an example, an energy resolution of $\Delta E = 91$ keV FWHM was achieved for 20.7 MeV $^{238}$U ions, unmatched by any ionization based detector. This unique performance has potential advantage for energy registration of fission fragments for which, to our knowledge, CLTD's were never applied before.

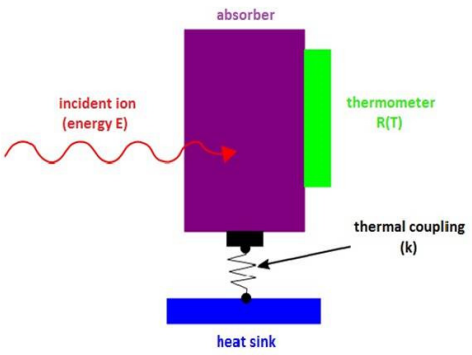


**Fig. 1.** Principle of particle detection with a calorimetric low temperature detector CLTD (for discussion see text).

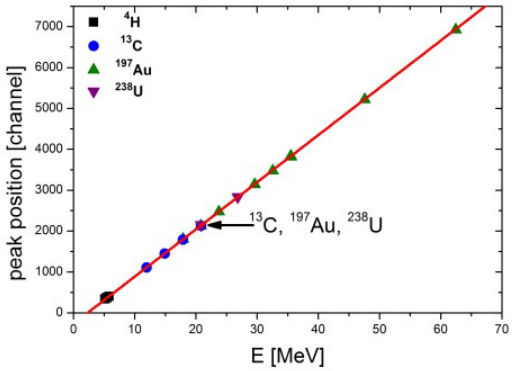


**Fig. 2.** Summary of a systematic study of the linearity of the energy response for various ions and energies. The solid line represents a linear fit to the data [9].

The detector array used in the present experiment consists of 25 pixels with a total active area of 15x15 $mm^2$ (fig. 3). To operate the array, a windowless pumped $^4$He-bath cryostat is used. An individual pixel is made of a sapphire substrate ($Al_2O_3$) serving as absorber with a thickness of 430 µm and an area of 3x3 $mm^2$. On its rear side, an aluminium thermistor is evaporated as temperature sensor, operated at the phase transition between the normal and superconducting state at $T_C$ @ 1.5 K. Proper thermal coupling is provided to a heat sink. The insert in fig. 3 (fig. 3b) shows a single pixel, with an additional gold wire strip serving as heater used for temperature stabilization. A weak $^{241}$Am α source is facing the array for calibration purposes. For the readout, conventional pulse electronics consisting of low-noise preamplifiers and flash-ADCs are used.

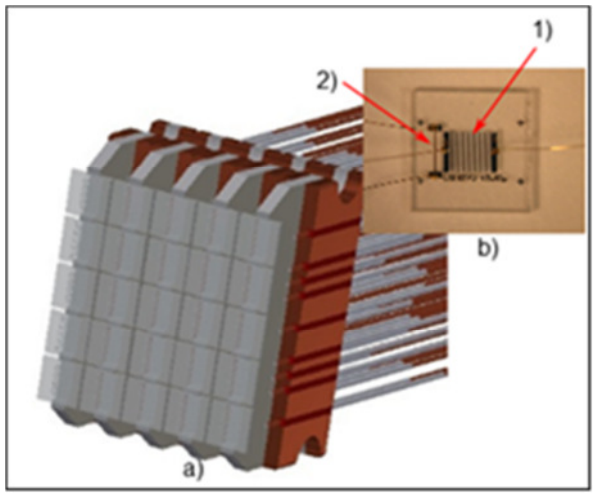


**Fig. 3.** a) Design of the CLTD-array consisting of 5 x 5 detector pixels.
b) 3x3$mm^2$ wide single pixel with Al-transition-edge sensor (1) and Au-heater (2).

## 3. Experimental set-up at LOHENGRIN

The $^4$He-bath cryostat containing the CLTD array of 25 independent detectors with a total active area of 15x15$mm^2$ was coupled, without any entrance foil, to the straight exit flange of LOHENGRIN inclined by 35° (see figs.4,5). However, in a first attempt, with positioning the $Si_3N_4$ absorbers on a movable manipulator at a distance of 95 cm outside the cryostat, the adaption of the cryogenic system to the LOHENGRIN beam line affected the CLTD energy resolution by heat radiation and, accordingly, required a small entrance slit of 30 $mm^2$. Also, having the $Si_3N_4$ absorbers upstream, largely reduced counting efficiency due to small-angle scattering and increased the background by contaminating mass lines. To counter these problems, for the second run a series of $Si_3N_4$ foils with a total thickness of 4.4 µm were placed inside the cryostat 9 cm ahead of the CLTDs. Heat absorption of these $Si_3N_4$ foils improved the thermal stability of the detectors and enabled an entrance slit as large as the size of the absorber foils (16x10 $mm^2$). Counting efficiency accordingly improved by a factor of 20. Optionally, some more absorber foils were still mounted onto the manipulator for varying the total absorber thickness up to 6.5 µm, at the cost of reduced transmission. A silicon PIN-diode, not shown in fig. 5, which served as a yield monitor for adjusting the LOHENGRIN mass separator was also mounted on the manipulator. Finally, towards the end of the experiment, the absorber configuration was again modified, with the 6.5 µm foil stack close to the detectors.

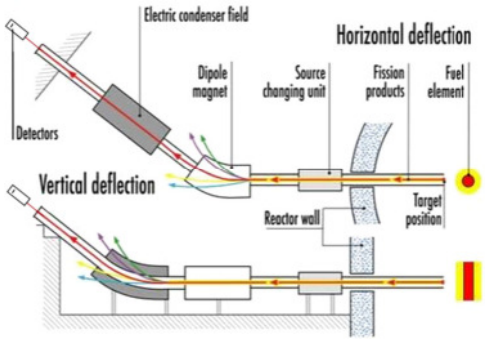


**Fig. 4.** Sketch of the LOHENGRIN recoil separator showing the flight path of the fission fragments emitted from the target close to the reactor core.

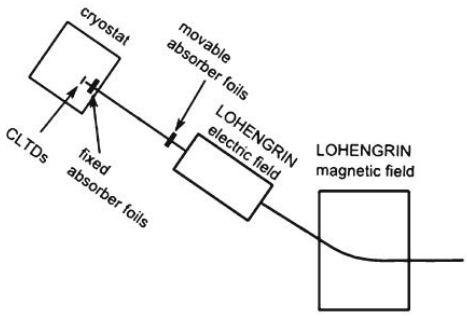


**Fig. 5.** Schematic view of the LOHENGRIN with the arrangement of silicon nitride absorber foils and the CLTD's as residual-energy detector.

## 4. Experimental results and discussion

The quality of nuclear charge separation was studied for selected masses in the region $82 \leq A \leq 132$ as a function of degrader thickness (about 4 to 8 µm of $Si_3N_4$) and for fission fragment kinetic energies from 55 to 110 MeV. For the light fragment group (Z<43, A<107), good Z-resolution was obtained (see fig. 6), sufficiently high to clearly separate individual charges in the residual-energy spectra and, as expected, with the resolution power improving towards higher energy losses. We could already match the historically best Z-resolutions, e.g. $Z/\Delta Z = 51$ at Z=37 ($\Delta Z$ is defined as the ratio of the peak width in FWHM and the difference between adjacent peaks in the rest-energy spectra), achieved conventionally with Parylene-C absorbers and ionization chambers [14, 15].

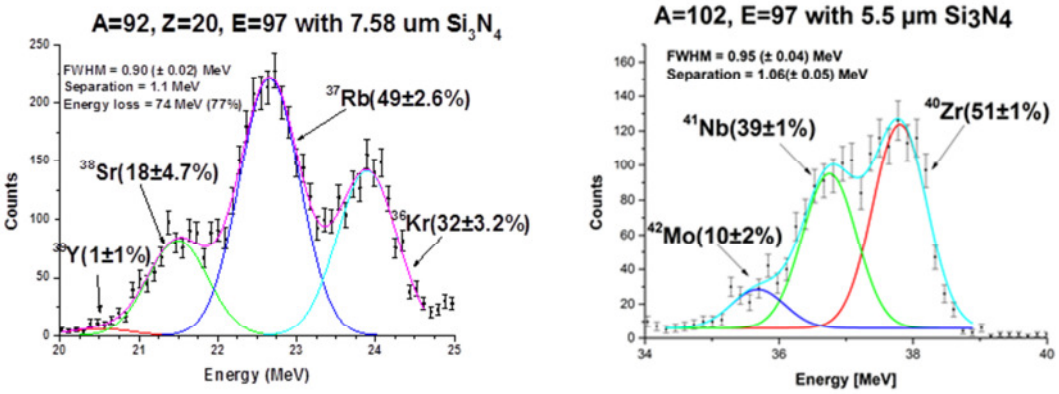


Fig. 6. Residual energy spectra for fission fragments from $^{235}U(n_{th},f)$. (a) A = 92, E = 97 MeV with 7.6 µm of $Si_3N_4$. (b) A = 102, E = 97 MeV with 5.5 µm of $Si_3N_4$.

For a precise determination of fractional independent fragment yields with LOHENGRIN it is mandatory to measure systematically 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 for extracting fragment yields is still in progress, and all data shown at present are considered as preliminary. Extended measurements were performed for mass A = 92, for various ionic charges ($q$ = 17, 19, 20, 21, 25) and energies ($E$ = 77, 84, 92, 97, 102 MeV), and partly for masses 106–109. The isotopic fission yield

of $^{92}$Rb from $^{235}$U($n_{th}$, f) was of particular interest, since more precise data than available [16] have been requested recently for achieving a better understanding of the reactor antineutrino spectrum [17,18]. The decay of this isotope, along with $^{96}$Y, is a main contributor to the integral antineutrino spectra above 4 MeV that are probed by sterile neutrino searches.

The investigation of isotopic yields for the heavier masses A > 106 (fig. 7) was started with the aim to study the onset of even-odd effects in the transition region from the light fragment group towards the symmetry region, which is of high interest for verification of nuclear models [19]. The data analysis concerning these fragment yields is presently in progress.

For heavy fragments the obtained Z-resolution was insufficient to resolve individual lines in the residual energy spectra (see fig.7 b). Nevertheless, the measured energy distributions show asymmetric shapes that enable a Z-yield determination, if parameters like peak width and energy loss difference of adjacent Z could be fixed. Different fits to the measured energy distributions, assuming 2 or 3 contributing charges or fixing the fractional yields using JEFF tables, give resolutions of Z/ΔZ=32-39 for Z=51, demonstrating a significant improvement in this mass and energy range as compared to previous measurements [15].

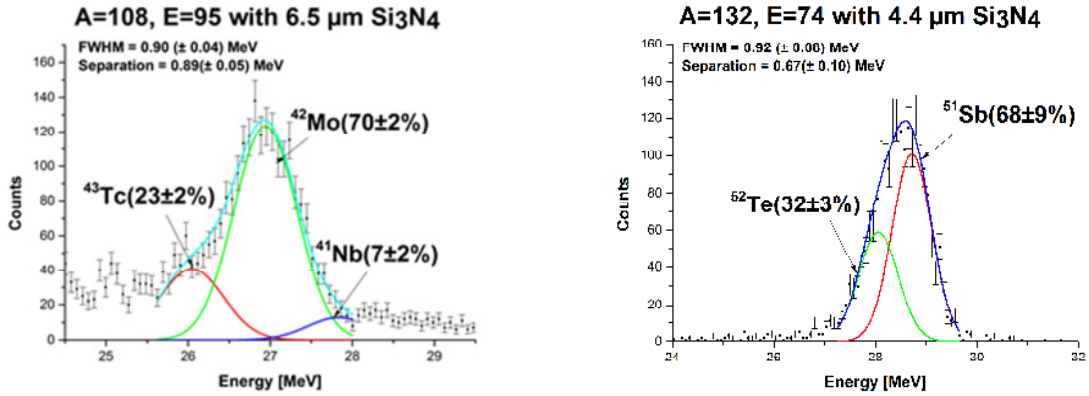


Fig. 7. Residual energy spectra for fission fragments from 235U($n_{th}$,f). (a) A =108, E = 95 MeV with 6.5 µm of $Si_3N_4$. (b) A = 132, E = 74 MeV with 4.4 µm of $Si_3N_4$.

## 5. Summary and outlook

The passive absorber method, for the first time with using $Si_3N_4$ membranes as degraders and CLTD's as residual-energy detectors, was successfully applied at the LOHENGRIN mass separator at the ILL for the investigation of isotopic yields of fission fragments. A systematic study on *Z* resolution with various degrader arrangements was performed in a wide range of fragment masses A and energies E.

For light fission fragments (Z < 43, A <107) we could already match the historically best Z-resolutions achieved conventionally with Parylene-C absorbers and ionization chambers [4,14,15]. Due to recent nuclear physics interest on new data for the isotopic yield of $^{92}$Rb, the present system was used for systematic charge-yield measurements on A = 92. Particularly in the case of $^{235}$U, two nuclei stand out for the high-energy region of the antineutrino spectrum emerging from nuclear power plants, $^{92}$Rb and $^{96}$Y. This is due to a combination of a large cumulative fission yield, a large $\beta-$ *Q* value and a large ground-state to ground-state $\beta$- feeding intensity. Currently, the $^{96}$Y yield is under study also.

In the heavier mass region, we obtained significant improvement as compared to previous setups, but the promising results from our test experiment at the MLL tandem accelerator could not be reached up to now. Therefore, it was not possible to achieve all our aims which were assessed on the basis of the results from the test experiment at MLL.

In the meanwhile, the experimental set-up was upgraded by installing a remotely controlled sample changer for $Si_3N_4$ absorber foils inside the cryostat, at a short distance to the CLTDs.

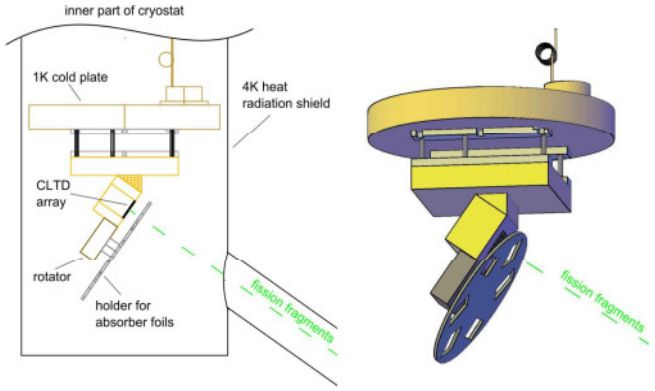


Fig. 8. Design of the new configuration of the CLTD array and the movable absorber foil system inside the cryostat.

The design of the new system is displayed in fig. 8. The rotatable disk, with six positions for $Si_3N_4$ foils of 16 x 10 $mm^2$ area, is operated by a remote-controlled piezo-driven rotary stepper positioner. We use the system ANR240/RES from Attocube [20], which consists of a positioner with resistive encoder along with the controller ANC350. The device operates under vacuum and at temperatures below 1°K, and allows a reproducible positioning with an accuracy of 0.050 ° (≈ 30 µm for the current design). It was already proven in the last run that $Si_3N_4$ foils survive the cooling down and warming up procedures. In addition, the CLTD array will be tilted perpendicular to the beam, increasing the effective detector area by 20% as compared to the previous runs. In a running experiment these modification has already shown considerable improvements with respect to transmission, resolution and flexibility for measurements in different mass and energy ranges.

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