Possible anisotropy in the emission of fission fragments

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Abstract
This study on $^{234}$U(n,f) focused on the vibrational resonance at the incident neutron energy $E_{\text{n}}=770$ keV. Due to the strong angular anisotropy, fluctuations of the fission fragment (FF) properties were predicted. The bipolar angular anisotropy was verified in this work and a possible new correlation to anisotropic FF emission has been observed. The mass distribution was found to have the biggest difference in asymmetry, at the vibrational resonance and was less asymmetric in emission along the axis of the beam direction. A corresponding anisotropy in the total kinetic energy was also observed. The observed effect was consistent with the change in the mass distribution. At last, the experimental data were fitted based on the Multi-Modal Random Neck-Rupture (MMRNR) model. The yield of the standard-1 mode was found to increase at the resonance.

1 Introduction
The motivation for this study on $^{234}$U(n,f) is the apparent need for nuclear data concerning this reaction. In addition to the importance of these data for nuclear applications, it can also be used to review parts of the modelling of fundamental fission dynamics due to the interesting properties of $^{234}$U(n,f). Several works, e.g. Refs. [1–3], investigated the anisotropic fragment emission in $^{234}$U(n,f) which is due to a prominent vibrational resonance in the sub-barrier region, at $E_{\text{n}}=770$ keV. However, to our knowledge, only one measurement exists on the energy distributions for $^{234}$U(n,f) [4] and no measurement is available on the mass distribution.

2 Background
The angular anisotropy in fission has been well studied. However much less is known on possible correlations with other fission-fragment observables like the mass distribution. A few works suggested an angular-anisotropy dependence of the fragment masses [5–8], and others disproved these findings e.g. [9–12]. Classical models favour an anisotropy independent on the mass [13]. The reason can be found in the fission barrier, which is responsible for the angular distribution according to the theory of Bohr [14]. The fission barrier height, is assumed to be the same for all asymmetric fission events, therefore the FF angular distribution, is also assumed to be the same [8, 10]. In modern fission models, for instance the Multi-Modal Random Neck-Rupture model (MM-RNR), the angular anisotropy may be mass dependent. Basically, each fission mode which has its own mass distribution, could in fact have different angular distributions [15, 16]. In this study we searched for possible correlations between the mass distribution and the prominent angular anisotropy which is peaked at 835 keV incident neutron energy. Moreover, we performed a fission mode analysis to understand the possible influence on the fission observables.

3 Experiments
In total 14 different measurements were collected at the incident energies: 0.2, 0.35, 0.5, 0.64, 0.77, 0.835, 0.9, 1.0, 1.5, 2.0, 2.5, 3.0, 4.0 and 5.0 MeV. The experiment is performed using neutrons from
the 7 MV Van de Graaff accelerator (MONNET) at the Institute for Reference Materials and Measurements (IRMM) in Geel, Belgium. For FF detection, a Twin Frisch Grid Ionization Chamber (TFGIC) was utilized. It has two anodes, two Frisch grids and one common cathode (see Ref. [17] for more experimental details). The sample used for irradiation is a $^{234}$UF$_4$ target enriched to 99% $^{234}$U and placed in the center of the cathode. P-10 counting gas was used in the chamber at a pressure of 1.05 $\times$ 10$^5$ Pa. By measuring the FF pulse height (proportional to the FF energy) and the emission angle, the pre-neutron emission masses could be calculated based on conservation of energy and momentum. A digital data acquisition system was used to store the raw signals from the charge sensitive pre-amplifiers. Several advantages were achieved by applying the digital techniques compared to conventional analogue techniques e.g. improving the angular resolution, verifying the correct grid-inefficiency correction and successfully correcting for $\alpha$ pile-up [17–19].

4 Analysis

The analysis of the data is based on the 2E method. As an absolute energy calibration, $^{235}$U(n$_{th}$,f) was measured with the same setup. Well known literature data on TKE and $\langle A_H \rangle$ were used for this reaction in the calibration. The angular resolution is reduced at higher emission angles due to the energy losses in the sample, therefore only events with $\cos(\theta) \geq 0.5$ were selected for the FF mass calculation. The analysis took into account the correction for the pulse-height defect, neutron-momentum transfer and the energy-losses in the sample. One crucial step in the analysis is the neutron multiplicity $\nu(A,TKE,E_n)$. Since it was not measured in this experiment, $\nu$ had to be parametrized based on data from neighbouring uranium isotopes, $^{233}$U(n,f) and $^{235}$U(n,f) [20]. The TKE dependence of $\nu(A,TKE)$ was parametrized as in Ref. [21] and the dependency on incident neutron energy was also corrected for using available data on $\nu_{tot}(E_n)$ for $^{234}$U(n,f) [22]. The angular anisotropy was calculated relative to the supposed isotropic thermal fission of $^{235}$U. The angular distributions were fitted in the center-of-mass system, with Legendre polynomials. The fit range was set to $0.3 < \cos(\theta) < 0.9$ due to the degrading resolution outside this range.

5 Results

The angular anisotropy found in Refs. [1–3] were confirmed in our work. The maximum anisotropy at the vibrational resonance was peaking at $E_n=835$ keV and had a minimum at $E_n=500$ keV. The changes in TKE were however different to the previous measurement. The TKE as a function of neutron energy increases at the resonance, contrary to the findings of Ref. [4]. Several attempts were made to understand the possible reasons behind this difference. We now believe that the difference in solid angle coverage may be the reason since only a small angle coverage was allowed when using surface barrier detectors, as in Ref. [4]. We found that the high TKE events contributing to the increasing TKE at the resonance, originate from events with higher emission angles. In fact, near 0° (relative to the incoming beam) the measurement at the resonance energy showed a slightly lower TKE than outside of the resonance energy, as observed in Ref. [4]. In Ref. [4], two different geometries were used, at 0° and 90°, respectively. The TKE was higher for the 90° run, however, probably due to the strong angular anisotropy the interpretation was different.

This apparent angle-dependent TKE is not straight forward and needed better quantification. The change in TKE as a function of $\cos(\theta)$ is probably not linear but a linear fit was anyhow applied as an approximation and for simplicity. The resulting change in slope was striking, showing a clear trend in correlation to the main vibrational resonance. The difference in TKE, between 0° and 90°, was at highest for the fission in the resonance. A similar fit was performed, now on the mass distribution as a function of $\cos(\theta)$. The trend observed in the TKE must have a direct link to the mass distribution and a possible anisotropy there as well. Indeed, after plotting the slopes of the different fits, a clear trend showed a higher anisotropy in mass emission in correlation to the vibrational resonance at $E_n=770$ keV. The observed effect was a more symmetric mass distribution for higher emission angles. Since a
more symmetric yield distribution preferable leads to higher TKE, these findings are consistent with the changes in TKE at the resonance.

To further study this effect, fission mode parametrizations based on Ref. [15] were used to fit the two-dimensional TKE vs. mass distributions. The 3 modes used, standard-1, standard-2 and super-long describe the asymmetric, very asymmetric and symmetric fission divisions, respectively. The mode-weight analysis showed that the standard-1 fission mode is actually increasing at the vibrational resonance and since it is giving higher TKE values, it is consistent with the higher TKE found at the resonance. The combination of a possible anisotropic mass emission and the growing standard-1 yield at the vibrational resonance, could indicate an angle-dependent mode change. Since the mass distribution becomes more symmetric for higher emission angles, at the same time standard-1 increases and the TKE becomes larger, the mode weight change could be angular-anisotropic. If true, the two standard modes may have slightly different angular distributions. As discussed earlier based on the Bohr theory, the angular distributions are closely related to the barrier height. So, could this be a (first) evidence on a different barrier height for the two standard fission modes?

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References