Ternary Fission Induced by Polarized Neutrons

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Abstract. P-odd and T-odd asymmetries in the emission of fragments and light particles have been investigated in ternary fission induced by polarized neutrons. P-odd asymmetries unambiguously point to a violation of parity in the fission process. By contrast, T-odd asymmetries do not necessarily imply a violation of time reversal invariance. The asymmetries observed are rather due to a final state interaction between the light ternary particle and the nucleus from which they are ejected. New results with interesting information on the ternary fission process are presented.

INTRODUCTION

In the reaction called ternary fission, besides the two fission fragments proper, a third charged particle is emitted. At low excitation energies, i.e., in spontaneous and thermal neutron induced fission, the ternary process is quite rare. Typically the probability for ternary fission is only between 2·10⁻³ and 6·10⁻³ of binary fission. In most cases the ternary particle is a light nucleus with hydrogen and helium isotopes contributing the lions share with about 97% of the total ternary yield. Among these nuclei the ⁴He-isotope carries about 90% of the total yield and is hence by far the most abundant ternary particle. Besides the ternary particle yields, both the energy distributions and the angular distributions relative to the fission axis have been studied extensively. In particular, for spontaneous and thermal neutron induced fission comprehensive reviews are available [1, 2].

In the following we report on a series of experiments where ternary fission was explored following capture of cold or hot polarized neutrons. To reach statistically significant results intense polarized neutron fluxes are required. All experiments to be described were performed at the High Flux Reactor of the Institut Laue-Langevin in Grenoble, France. At this reactor both cold and hot neutron sources are installed providing intense neutron beams. The polarization of the neutron beam allows a search for P-odd and T-odd asymmetries and correlations in the angular distributions of fission fragments and ternary particles. Several fissile targets from uranium to curium were investigated.

LAYOUT OF EXPERIMENTS

The layout for all experiments to be discussed here was very similar. A typical setup is sketched in Fig. 1. The targets were mounted in the centre of a reaction chamber and irradiated by a polarized neutron beam running horizontally. In the figure the beam polarization is shown to be aligned in or opposite to the beam direction. The spin was flipped periodically and data were taken alternatively for the two spin orientations. This common spin flip technique allows to get rid of most of the systematic errors. The target isotopes under study were ²³³U, ²³⁵U and ²³⁹Pu. Typically a total amount of a few mg was evaporated onto a thin Ti foil (~100 µg/cm²) that was transparent to fission fragments. The specific thickness of the target material was also ~100 µg/cm². In some experiments, in particular with ²³⁹Pu as the target, a thick and non-transparent backing had to be used for safety reasons. The fission fragments were detected by multiwire proportional counters (MWPC) placed in Fig. 1 to the left and right. They were operated with CF₄ as the counting gas at low pressure (~15 mb). The MWPCs were position sensitive with a
resolution of better than 1mm in both coordinates. This allowed reconstruction of the location on the target where fission took place. The light particles were intercepted by silicon PIN diodes. Two arrays with up to 20 diodes each were positioned at right angles to both, the neutron beam and the average fission axis defined by the common axis of the MWPCs. Since ternary particles are emitted predominantly near a plane perpendicular to the fission axis, the geometry chosen for the position of detectors optimizes the sensitivity for ternary fission. The size of the diodes was 3x3 cm². By inspecting the signal risetime in the diodes a particle identification was achieved [3]. The H-isotopes were well separated from the α-particles. To avoid radiation damage by fission fragments and α’s from radioactivity, the diodes had to be covered by a thin protection foil made from aluminum. The foil introduces, however, a low energy cut-off in the energy spectra of the ternary particles.

FIGURE 1. Sketch of the beam-target-detector arrangement.

Asymmetries in the angular distributions of fragments have to refer to specific fragments. Since fragments from fission fly in opposite directions, asymmetries change sign when referred to one or the other fragment. The minimum requirement, hence, is that fragments from the light and heavy mass group can be discriminated. In the present experiments this was achieved by evaluating the times of flight for the fragments and ternary particles, knowing the location of the nucleus having undergone fission.

For the cold neutron beam with an average neutron energy of ~4 meV, the equivalent thermal flux was ~ 6·10⁸ n/(cm²s) with a polarization of 95%. Depending on the amount of target material employed, the count rates for binary fission were on average 5·10⁹/s for ternary fission 2·10⁷/s. The hot beam had an average neutron energy of 0.16 eV, a polarized flux of ~1·10⁴ n/(cm²s) at a polarization of 82%. To compensate for the lower flux and fission cross section the amount of fissile material (²³⁵U) had to be increased to 15 mg on a non-transparent Al backing.

P-ODD ASYMMETRIES

Searching for P-odd asymmetries in the angular distributions of fragments from fission reactions induced by polarized neutrons an appropriate observable is the scalar product \( \sigma_n \cdot p_{LF} \) with \( \sigma_n \) the spin of the neutron and \( p_{LF} \) the momentum of the light fragment. Both vectors have unit length. Note that it is by convention that the observable is referred to the light fragment LF. As a pseudo scalar \( \sigma_n \cdot p_{LF} \) is P-odd and as a rule, whenever a pseudo scalar is found in nature, parity is violated.

For optimum efficiency the spin \( \sigma_n \) has to be aligned parallel to fragment momentum \( p_{LF} \) in experiments probing parity violation. In the experimental layout of Fig. 1 this means that the spin has to be turned from the direction shown by 90° to face the MWPC detectors. The angular asymmetry of fission fragments to be inspected then is

\[
W(p_{LF})d\Omega_{LF} \sim [1 + \alpha_{PNC}(\sigma_n \cdot p_{LF})]d\Omega_{LF} \tag{1}
\]

The coefficient \( \alpha_{PNC} \) is a measure of the size of parity non-conservation (PNC). A non-zero PNC effect was indeed discovered in the late seventies in binary fission [4]. It came as a surprise. But quite promptly theoretical models were proposed which could explain the experimental results [5, 6]. In these models the crucial point is that e.g., following the capture of an s-neutron there is an albeit small probability that the weak interaction will mix s- and p-states having the same total angular momentum \( j \). It has to be stressed that this mixing occurs at the compound nuclear stage of the fission process.

In later experiments PNC was also studied for ternary fission reactions [7, 8]. In these studies again the angular asymmetry of fragments is investigated but, in addition, the presence of a ternary LCP is required. The best studied reaction is \( ^{233}U(n_{th},f) \). The most recent results for this reaction are

\[
<\alpha_{PNC}^{\text{bin}}> = + 0.400(17) \cdot 10^{-3} \tag{2}
\]

in binary fission [9] and

\[
<\alpha_{PNC}^{\text{tern}}> = + 0.37(10) \cdot 10^{-3} \tag{3}
\]

in ternary fission accompanied by an α-particle [10]. The averages in Eqs. (2) and (3) are taken over
fragment masses and kinetic energies and \( \alpha \)-particle energies. Within statistics the two results are identical. It is remarkable that the outcome of the PNC effect as seen in the angular asymmetry of fragments is the same whether an \( \alpha \)-particle is present or not. Without going into details here, it should be added that similar results were obtained for the left-right asymmetry of fragment angular distributions. Also there the size of the asymmetry is virtually the same in binary and ternary fission.

It is instructive to inspect the PNC effects not only as averages as in Eqs. (2) and (3) but more in detail as a function of fragment and LCP properties. Again for the \( ^{235}\text{U}(n_{\text{th}},f) \) reaction it could be shown in separate studies that the size of the PNC effect does not vary with mass and/or energy of the fragments [9]. The limited statistics does not allow to perform this type of analysis also for ternary fission. However, the dependence of the PNC asymmetry of fragments on the \( \alpha \)-particle energy could be determined [10]. The results are displayed in Fig. 2. The statistical error bars are large but anyhow there is no evidence for any strong dependence of the PNC effect on \( \alpha \)-energy.

We note in passing that an analogous PNC effect may also be analyzed for the LCP particles by replacing in Eq. (1) the fragment momentum by the LCP momentum. Once more for the \( ^{235}\text{U}(n_{\text{th}},f) \) reaction, an upper limit of \( 0.16 \cdot 10^{-3} \) was measured. Hence, the PNC effect for the LCPs is definitely smaller than for the fragments [see Eqs. (2) and (3)].

The experiments on PNC effects in ternary fission allow one to draw conclusions giving interesting insights into the process. According to a generally accepted theory of A. Bohr [12], for fission at low excitation energies the angular distributions of fission fragments are settled at the saddle point. The spinning top wavefunctions \( D_{3\text{M}}^\text{Y} \) of the transition states at the saddle point characterize the distributions. Parity violating angular distributions should make no exception to this rule. The equality of the PNC effect in binary and ternary fission as expressed in Eqs. (2) and (3) is, hence, interpreted to imply that up to the saddle point the two decay modes, binary and ternary fission, are described by identical wavefunctions. The \( \alpha \)-particles, and probably also all other LCPs, become palpable only at a later stage of the fission process, i.e., close to or right at scission. The fact that the size of the PNC effect is not dependent on \( \alpha \)-energy (see Fig. 2) corroborates this view. Indeed, following A. Bohr's theory PNC asymmetries are frozen in at the saddle point and details of the scission configurations are not relevant. On the other hand, angular distributions and energies of ternary particles are generally accepted to be conditioned by the scission configuration. Therefore the PNC effect should not exhibit any dependence on \( \alpha \)-energy, as found experimentally. Finally, also the observation that the PNC effect for the \( \alpha \)-particles is much smaller than for the fragments is evidence for a late appearance of the LCPs. Loosely speaking the \( \alpha \)-particles are born too late to experience the influence of the weak interaction. The above ideas have been expounded already some time ago [13] but it is only more recently that all experimental evidence could be collected.

The notion of a late birth of the ternary particles should be seen together with the information on their birthplace, which is inferred from the pattern of angular distributions observed for the LCPs. From numerous studies it emerges that by far most of all \( \alpha \)-particles are emitted at roughly right angles to the fission axis, which by convention is approximated in experiments as the direction of flight of the light fragment [1, 2]. These are the so-called equatorial LCPs under study in the present experiments. Trajectory calculations supporting intuitive supposition establish that the ternary particles are ejected from the region of the neck that is forming between the two main fragments when these start to separate. The experimental information on the time and location when and where ternary particles come into existence gives credit to theories advocating double random neck rupture as the source of LCPs [14, 15].

**T-ODD CORRELATION**

The study of parity violation in fission having a long tradition, the idea came up whether not also the violation of time reversal invariance could be investigated. In analogy to the decay of polarized free neutrons it was proposed that a triple correlation (TRI) could be analyzed in ternary fission that is T-odd [16]. The observable in question is \( \sigma_* \cdot (p_L \times p_T) \) with the notations \( \sigma_* \) and \( p_L \) being the momentum of the ternary particle. All vectors are understood to have unit length. While

![FIGURE 2. PNC asymmetry of fragments in \( \alpha \)-accompanied ternary fission of \( ^{235}\text{U}(n_{\text{th}},f) \) as a function of \( \alpha \)-energy from 10 to 30 MeV.](image-url)
being T-odd the observable is P-even. The angular correlation to be explored in experiment reads

\[ W(p_{LF}, p_{TP})d\Omega_{p_{LF}}d\Omega_{p_{TP}} \]

\[ \sim (1 + D \sigma_n [p_{LF} \times p_{TP}] )d\Omega_{p_{LF}}d\Omega_{p_{TP}}, \quad (4) \]

where the coefficient D indicates the size of the correlation. It should be stressed that, unlike the PNC measurements in ternary fission, in the TRI correlation of Eq. (4) besides the neutron spin both the fragment and the ternary particle momentum enter.

The layout of TRI experiments is sketched in Fig. 1. For optimum sensitivity the neutron spin has to be aligned perpendicular to the plane defined by the two momentum vectors \( p_{LF} \) and \( p_{TP} \) as shown in the figure. The correlation was surveyed for several thermal neutron induced fission reactions in the actinides. Results for the TRI coefficient D are summarized in Table 1 with \( \alpha \)-particles as the LCPs:

<table>
<thead>
<tr>
<th>Target</th>
<th>( ^{233}\text{U} )</th>
<th>( ^{235}\text{U} )</th>
<th>( ^{239}\text{Pu} )</th>
<th>( ^{245}\text{Cm} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \langle D \rangle \times 10^3 )</td>
<td>-3.9(1)</td>
<td>+0.8(1)</td>
<td>-0.2(3)</td>
<td>-1.3(5)</td>
</tr>
<tr>
<td>( I^\pi )</td>
<td>5/2( ^{-} )</td>
<td>7/2( ^{+} )</td>
<td>1/2( ^{-} )</td>
<td>7/2( ^{+} )</td>
</tr>
</tbody>
</table>

For the \( ^{233}\text{U}(n_{th},f) \) reaction the TRI correlation could also be determined for the H-isotopes (mostly tritons). The result \( \langle D \rangle = -2.9(5) \times 10^{-3} \) possibly indicates that the TRI correlation depends on the particle type being set free as LCP. A further result was obtained replacing the cold neutrons by hot neutrons. Again for \( ^{233}\text{U} \) as the target, a TRI correlation with the size \( \langle D \rangle = -2.4(8) \times 10^{-3} \) was measured at a neutron energy of 160 meV.

As in the case of PNC asymmetries further information should be gained by scanning the TRI correlation as a function of fragment and ternary particle properties like masses and energies. Unfortunately the mass and/or energy resolution of the MWPC detectors was barely sufficient to find unequivocal results. However, the dependence of the TRI coefficient on \( \alpha \)-particle energy could be determined with good resolution. This dependence is shown in the figures below for the reaction \( ^{233}\text{U}(n_{th},f) \).

The \( \alpha \)-energy spectrum is reproduced for orientation in Fig. 3. The low energy part of the spectrum is cut off at about 7.5 MeV due to the energy loss in the Al foils protecting the PIN diodes. In Fig. 4 the modulus \( |D| \) of the TRI correlation is plotted. In contrast to the behavior of the PNC effect in Fig. 2, the strong increase of the TRI correlation with \( \alpha \)-energy is conspicuous. At the high energy limit of the distribution the correlation comes close to \( 10^{-2} \).
be expected to be contingent on the details of the scission configuration (see Fig. 2 for the PNC effect). Yet, for the TRI correlation precisely this dependence is observed in Fig. 4. The conclusion is that we are facing a final state interaction.

Next one has to ask which type of final state interaction could be made responsible for the TRI correlation observed. To answer this question it is helpful to resuffle the vectors in the TRI observable:

\[ \sigma_n \cdot [p_{LF} \times p_{TP}] = p_{TP} \cdot [\sigma_n \times p_{LF}] \]  

(5)

Referring to Fig. 1, the expression on the RHS of Eq. (5) describes the TRI correlation as an asymmetry in the emission of the ternary particle \( p_{TP} \) with respect to a plane defined by the neutron spin \( \sigma_n \) and the momentum of the light fission fragment \( p_{LF} \). In the detector configuration of Fig. 1 this means that, depending on the neutron spin orientation, the ternary particles are emitted preferentially upward or downward.

Several conjectures have been put forward to explain the above asymmetry. The most appealing models invoke a Coriolis interaction. While detailed numerical results in a microscopic model of fission are still pending [18], a statistical model version gives a reasonable account of the TRI correlation [19].

Before expounding this latter model let us first try to demonstrate the impact of the Coriolis force in a pedestrian approach. The sketch in Fig. 5 illustrates the idea. When a nucleus with spin \( I \) captures an \( s \)-neutron, the compound nucleus will have the spin \( J = I \pm 1/2 \). For example, in the compound nucleus \( ^{234}U \) the spin will be 2° or 3°. Depending on the capture state the nuclear spin \( J \) will be either oriented parallel or anti-parallel to the neutron spin. In any case, when flipping the neutron spin also the nuclear spin is flipped. In Fig. 5 a neutron spin orientation is assumed corresponding to the experimental situation in Fig. 1.

In thermal neutron fission of \( ^{235}U \) the accessible states at the saddle point lie in the single particle excitation gap and are, hence, collective in character. In the spirit of A. Bohr's theory [12] the states will retain this character down to the scission point. The scissioning nucleus has, therefore, to be viewed as a rotor with spin \( J \), which in ternary fission is emitting a particle with spin \( j \). In case of an \( \alpha \)-particle the spin \( j \) is just the orbital momentum \( \alpha \). Coming to the Coriolis interaction, in semi-classical approximation the Coriolis Hamiltonian is \( H_{tor} = -\hbar \omega \cdot j \) with \( \omega \) the angular frequency of the rotor where \( \omega \approx \omega_r \). Therefore one has \( H_{tor} \sim J \cdot \alpha \) and as seen from Fig. 4 the Coriolis interaction is operative.

In classical language the inertial forces having to be taken into account in a rotating system are the Coriolis force \( F_{cor} = 2m(v \times \omega) \) and the centrifugal force \( F_{cen} = m\omega \times (r \times \omega) \), with \( m \) the mass, \( v \) the velocity and \( r \) the location of the particle to be emitted. Though these two forces are minute compared to the Coulomb force exerted by the two main fragments on the ternary particle, they are seen in the figure to add constructively for particle emission in the direction of rotation, while for emission opposite to the direction of rotation they add destructively because \( F_{cor} \) is inverted but \( F_{cen} \) not. It is hence concluded that \( \alpha \)-particle emission is favored in the direction of motion of the rotor, i.e., the case shown in the figure. Finally it has to be recalled that the direction of rotation is steered by the nuclear spin \( J \) and in experiment ultimately by the neutron spin \( \sigma_n \). It is thus seen that the Coriolis interaction can give rise to the TRI correlations observed.

Quantitative predictions were derived in a statistical model [19]. A key ingredient of the model is a level density formula taking explicitly into account collective angular momentum. The formula for the TRI correlation reads

\[ D = \frac{h^2}{4\pi^2} \cdot \frac{J_{\alpha}}{2\Theta} \cdot \frac{\sqrt{a}}{\sqrt{E_{sci}}} \cdot |\mu \cdot P(J)|, \]  

(6)

with \( \Theta \) the moment of inertia, \( a \) the level density parameter, \( E_{sci} \) the intrinsic excitation energy at scission, \( \mu \) the polarization transfer from the compound nucleus to the fragments and \( P(J) \) the polarization of the compound nucleus. In Eq. (6) the factor \( J_{\alpha} \) is typical for the Coriolis interaction. The model gives the correct order of magnitude of \( 10^{-3} \) for the TRI coefficient \( D \). Several features of Eq. (6) can be checked out in experiment.
With $D \sim J$ the TRI correlation should be nil for compound spins $J = 0$. An example is provided by the $^{239}$Pu(n$_{th}$,f) reaction which was studied recently. For thermal (or even cold) neutrons the $0^+$ state is dominating capture and, in fact, as to be read from Table 1 the TRI correlation is compatible with zero.

The observed $\alpha$-energy dependence of the TRI correlation (see Fig. 4) is hidden in the excitation energy $E^*_{sci}$ of the formula for $D$ in Eq. (6). There is, unfortunately, no direct experimental evidence or a theoretical estimate available for this dependence. What is known from experiment is that not surprisingly the total excitation energy, i.e., the sum of deformation and intrinsic energy at scission decreases linearly with increasing $\alpha$-energy. The same behavior is assumed to obtain for the intrinsic excitation energy $E^*_{sci}$. To find the linear relation between $E^*_{sci}$ and $E_\alpha$ it is first recalled that the average intrinsic excitation at scission has been determined in studies of even-odd effects in the charge yields of fragments. According to the reasoning in the section on parity asymmetries, this average intrinsic excitation energy should be the same in binary and ternary fission. It appears reasonable to presume that the average $<E^*_{sci}>$ will in ternary fission correlate with the average $<E^*_{sci}>$. It is further conjectured that cold fission with $E^*_{sci} = 0$ is attained for the maximum $\alpha$-energy observed. It is thus found that the requested relation reads $E^*_{sci} \approx (6 - 0.2 E_\alpha)$ per fragment, all energies being given in MeV. The function describes remarkably well the sharp increase of $|D|$ in Fig. 4 when the $\alpha$-energy approaches the upper limit of $E_\alpha \approx 30$ MeV.

For the PNC asymmetry theory invokes the interference of s- and p-waves of the incoming neutron. From experiments investigating the dependence of the asymmetry on neutron energy it is reported that for the reaction $^{235}$U(n,f) the PNC coefficient changes dramatically and even changes sign from $\alpha^{PNC} = + 0.4 \cdot 10^{-3}$ to $\alpha^{PNC} = - 0.6 \cdot 10^{-3}$ for neutron energies from thermal to about 160 meV [20]. The strong variation is attributed to a p-resonance in the fission cross section at $\approx 160$ meV. By contrast, in the models proposed for the TRI correlation, p-waves are of no importance at all. The contention could be checked in the present experiments in the $^{235}$U(n,f) reaction. As communicated in a former section, the TRI coefficient amounts to $D = - 3.9(1) \cdot 10^{-3}$ for cold neutrons, while for hot neutrons with energy 160 meV one finds $D = - 2.4(8) \cdot 10^{-3}$. The small shift does not reveal any influence of the p-resonance.

In summary it can be stated that models describing the TRI correlation as a final state interaction based on the Coriolis interaction account convincingly for all experimental observations made up to the present.

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**REFERENCES**