Alpha-particle emission in ternary fission

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Summary. — According to recent data on equatorial and polar \( \alpha \)-emission in the ternary fission of \( ^{252}\text{Cf} \), the necessary energy to be supplied to the fissioning nucleus is of the order of the energy of the double giant dipole resonance. We show that a cluster model of fission can explain the appearance of various very energy-rich processes in an early stage of fission.

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1. – Introduction

The phenomenon of \( \alpha \)-particle accompanied fission was discovered in 1946 after irradiation of uranium-loaded photographic plates with neutrons [1]. Emission of other light charged particles (LCP) in fission was later reported too, and ternary (or LCP accompanied) fission has been extensively studied. In all cases, the mass distribution of the fission products is asymmetric, as in binary fission. Although the energy released in ternary fission is less than in the binary case, its relative yield is high (3–6 \( \times \) 10\(^{-5} \)) but an \( \alpha \)-particle is emitted in \( \sim \) 90% of the ternary fissions.

The most salient, still unexplained, features of ternary fission are the following:

1) The alpha particles are emitted at almost 90 degrees of the fission axis. The angle \( \theta_{\alpha} \) formed by the emitted \( \alpha \) with the fission axis oriented towards the light fragment has a mean value of 83 degrees according to Heeg [2]. This emission, called «equatorial», is generally distinguished from the rather rare «polar» emission (see sect. 4). The mechanism of particle emission is still unknown. A description of the various theoretical approaches is given by Mutterer and Theobald [3].

2) The «equatorial» ternary alpha particles emitted in the fission of actinides are emitted with the same mean kinetic energy of 15.9 MeV in fissioning systems as different as \( ^{231}\text{Pa} + n \), \( ^{233}\text{U} + n \), \( ^{237}\text{Np} + n \), \( ^{239}\text{Pu} + n \), \( ^{241}\text{Am} + n \), \( ^{242}\text{Am} + n \), \( ^{244}\text{Am} + n \), or \( ^{256}\text{Cf} \) (s.f.), \( ^{252}\text{Cf} \) (s.f.), \( ^{256}\text{Fm} \) (s.f.), \( ^{257}\text{Fm} \) (s.f.). The FWHM of the energy distribution is almost the same, from 9.6 MeV for \( ^{235}\text{U} + n \) to 10.9 MeV for \( ^{256}\text{Cf} \) (s.f.). However, \( E_{\alpha} \) is found to be somewhat smaller than 15.9 MeV in fusion-fission reactions, and in these cases the FWHM of the energy distribution is often found to be smaller than in the fission of the actinides [3].
No approach has yet succeeded in giving a theoretical explanation of this situation. An attempt, inspired by the work of Halpern [4], has recently been formulated by Woestheinrich [5] for estimating the missing energy necessary for the emission of the various LCP’s.

In sect. 2 we shall try to determine the energy needed by alphas emitted in ternary fissions in which the heavy fragment is $^{132}$Sn. In sect. 3 and 4 we shall suggest a possible explanation of the emission of «equatorial» and «polar» ternary alphas based on a description of the initial dynamics of any asymmetrical fission [6] and on the idea that various energy-rich processes, such as giant resonances, necessarily play a role at the early stage of fission.

2. – Estimation of the energy necessary for the alpha emission in a ternary fission process leading to a $^{132}$Sn heavy fragment

The study of the fragment mass distribution of binary and ternary spontaneous fissions of $^{252}$Cf reveals an important fact: binary and ternary distributions coincide only for $A = 132$ [3].

We interpret this observation as an indication that, for $^{252}$Cf fissions leading to a heavy fragment such as $^{132}$Sn, the $\alpha$-particle is necessarily formed from the matter of the complementary fragment in binary fission, i.e. $^{120}$Cd.

Instead of the usual representation of binary and ternary fission,

$^{252}$Cf $\rightarrow ^{132}$Sn + $^{120}$Cd + 236.6 MeV,

$^{252}$Cf $\rightarrow ^{132}$Sn + $^{116}$Pd + $^4$He + 230.2 MeV,

we suppose that a $^{44}$S is formed in $^{252}$Cf according to

$^{252}$Cf $\rightarrow (^{208}$Pb-$^{44}$S ) + 108.7 MeV

and that rearrangement processes can occur in this ($^{208}$Pb-$^{44}$S) molecule, according to

$^{208}$Pb-$^{44}$S $\rightarrow ^{132}$Sn + $^{120}$Cd + 127.95(0.65) MeV,

$^{208}$Pb-$^{44}$S $\rightarrow ^{132}$Sn + $^{116}$Pd + $^4$He + 121(0.68) MeV.

We assume further that the energy released in (3) is stored in the ($^{208}$Pb-$^{44}$S) molecule, principally as vibration energy, but that the rearrangement reactions (4) or (5) can occur for high values of the vibration quantum number $v$.

Equations (4) and (5) permit an evaluation, using experimental mass data [7], of the energy necessary for the emission, apparently by $^{120}$Cd, of $\alpha$-particles having a kinetic energy of 15.9 MeV, the mean kinetic energy of ternary alphas of $^{252}$Cf. Such an $\alpha$-energy would correspond to a $Q_a$ ($^{120}$Cd) value of 15.9 (1 + 4/116) = 16.45 MeV.

However, mass tables [7] reveal a negative $Q_a$ ($^{120}$Cd) value, −6.45 MeV. Thus we are led to the intriguing question: where does the energy difference $\Delta Q = 16.45 - (-6.45) = +22.9$ MeV come from?
3. – An hypothesis on the origin of the energy needed for equatorial \( \alpha \)-emission

According to a recent proposal [8], a double giant dipole resonance (DGDR) could appear in the fissioning system (3) either as a repercussion of the rupture of the bond of the \({}^{132}\text{Sn}\) remnant (eqs. (4) and (5)) with its 76 valence nucleons in the collision \( {}^{208}\text{Pb}-{}^{44}\text{S} \) at high \( v \) quantum number, or as a Coulomb excitation of core and cluster at the closest distance of approach of core and cluster: proton and neutron phases oscillate out of phase in the direction of the fission axis.

This hypothesis explains first the emission at almost 90 degrees of the most abundant \( \alpha \)-particles, as a consequence of the transversality of the electric field associated with a linear vibration of charged particles.

The observed emission angle \( \theta_{\alpha} \) could result from the vector addition of the electric field orthogonal to the fission axis and of the force responsible for the motion of the valence nucleons from the \({}^{132}\text{Sn}\) core to the \({}^{44}\text{S}\) cluster, also in the direction of the fission axis. This hypothesis further explains how the fissioning system can acquire an energy as high as 22.9 MeV for the creation of 15.9 MeV-\( \alpha \)s. It is indeed well known that the mean energy of an isovector giant dipole resonance in a \({}^{252}\text{Cf}\) nucleus can be equal to \( \sim 13.2 \) MeV. Thus a two-phonon giant dipole resonance [9] can supply an energy of about 26.3 MeV [10]. DGDRs have recently been observed in heavy-ion reactions at very high energies. The \( 3\alpha \)-decay of such a DGDR created in Coulomb excitations has been observed [11].

However, a pertinent explanation needs to explain the emission of \( \alpha \)-particles and of much heavier LCPs. For this reason we have to ask: Is the DGDR imagined by Mouze [6] really the two-phonon giant resonance? Is it not rather the result of two single GDRs, one occurring in the \({}^{132}\text{Sn}\) remnant (or in a fragment having almost the mass \( A = 132 \)) and the other in the cluster, which receives the valence nucleons released by the \( {}^{208}\text{Pb} \) core? Indeed, the flow of these valence nucleons probably occurs between nuclei in which protons and neutrons simultaneously vibrate out of phase. This new question is an important one, because it is necessary to find an explanation not only for the emission of ternary \( \alpha \)-s, but also for the emission of much heavier «light» charged particles, for example \({}^{24}\text{Si}\), and thus for a much greater expense of energy. The assumption that not only a GDR but also a DGDR could occur both in the \({}^{132}\text{Sn}\) remnant (or in a fragment having almost the mass \( A = 132 \)) and in the cluster receiving the valence nucleons would explain that, instead of the \( \alpha \)-particles, emitted with a very high yield, many other LCPs can also be emitted, but with a much smaller yield.

Han Hongyin [12] has pointed out that the multiplicity \( \nu \) of the prompt neutrons emitted by \({}^{252}\text{Cf}\) varies, as a function of the energy of the \( \alpha \)-s measured in coincidence, according to the law \( d\nu/dE_{\alpha} = -0.038 \, n/\text{MeV} \) around the mean value \( \nu = 3.13 \). This interesting observation suggests that perhaps a small part of the prompt neutrons could be the result of giant dipole resonances.

4. – An hypothesis on the origin of the energy needed for polar \( \alpha \)-emission

4.1. Remarks on the role of spherical clusters such as \({}^{132}\text{Sn}\) in asymmetric fission.

Besides the occurrence of GDRs and DGDRs in energy-rich rearrangement reactions of the primordial diclusteric molecule, the occurrence of giant monopole resonances in
processes such as eq. (4) or (5) has probably to be considered, because the release of as many as 76 valence nucleons by the \(^{132}\text{Sn}\) core (or of almost as great a number of such nucleons by somewhat heavier rumps) necessarily causes fierce repercussions in the rumps [13].

This hypothesis of the occurrence of giant monopole resonances could explain the appearance in the spontaneous fission of \(^{252}\text{Cf}\) of the so-called «non-statistical» \(\gamma\)-ray component reported by the Darmstadt-Heidelberg group [14].

The last proton shell which is closed in \(^{132}\text{Sn}\), at \(Z = 50\), is certainly the shell which suffers the strongest «shock» as a result of its stiffness. But the shell which is closed at \(Z = 58\), for example in \(^{140}\text{Ce}\), could still respond to the release of valence nucleons, even if its stiffness is considerably smaller.

Indeed Singer [15] has recently observed various peaks in the mass distribution of \(^{252}\text{Cf}\) fragments measured in coincidence with the \(\gamma\)-emission, not only at \(A \sim 131\), for \(E_\gamma \sim 6\) MeV but also at \(A \sim 140\), for \(E_\gamma < 1\) MeV.

The existence of this «non-statistical» \(\gamma\)-emission has been considered as doubtful by van der Ploeg [16]. However, we will show that the study devoted by Heeg to the emission of «polar» \(\alpha\)-particles in ternary fission provides an important new argument in favour of the role played in asymmetric fission by the doubly magic \(^{132}\text{Sn}\) nucleus—and thus in favour of Mouze’s interpretation of the «non-statistical» \(\gamma\)-ray emission. And this argument is important precisely because it is furnished not by the study of the \(\gamma\)-emission, but by the study of the \(\alpha\)-emission.

4.2. The «polar» alpha emission. – The «polar» emission along the fission axis has been discovered by Atneosen et al. [17], and further studied by Piasecki et al. [18]. This \(\alpha\)-emission occurs in less than 2% of the emissions of ternary alphas, and has a greater yield in the direction of the light fragment than in the direction of the heavy fragment. The mass distribution of the \(^{252}\text{Cf}\) fragments emitted in coincidence with the polar alphas, and their kinetic energy, have been measured by Heeg [2]. The following important facts need an explanation:

1) The mass yield shows, beside a broad distribution culminating at \(A \sim 140\), a distinct maximum at \(A \sim 117\) and \(A \sim 131\) having a narrower FWHM. This effect is clearly present for the alphas emitted in the direction of the light fragment, but can still be detected for the alphas emitted in the direction of the heavy fragment.

2) The kinetic energy \(E_f\) of one of the fragments (\(l\) or \(h\)) has been measured in coincidence with the emitted \(\alpha\)-particle not only in the case of equatorial emission, i.e. emission in a direction orthogonal to the fission axis (case noted \(eq\.), but also in the 2 cases of polar emission, i.e. either emission in the direction of the light fragment (case noted \(p.l\.), or emission in the direction of the heavy fragment (case noted \(p.h\.).

Now one finds that the fragment kinetic energy \(E_f\) for the light fragment has the same value for the case \(p.l\.) as for the case \(eq\.), whereas \(E_f\) for the heavy fragment is diminished by about 13 MeV for the case \(p.l\.) as compared to the case \(eq\.).

On the contrary, one finds that \(E_f\) for the light fragment is diminished by about 12 MeV for the case \(p.h\.) as compared to the case \(eq\.), whereas \(E_f\) for the heavy fragment seems increased by about 6 MeV for the case \(p.h\.) as compared to the case \(eq\.).

3) The energy distribution of the polar alphas is about the same for the case \(p.l\.) as for the case \(p.h\.). The mean value of the kinetic energy is in both cases \(\bar{E}_\alpha = 25\) MeV. This value is much greater than the mean value \(\bar{E}_\alpha = 15.9\) MeV of the equatorial alphas.
5. – Discussion and conclusion

These salient facts concerning the polar $\alpha$-emission show the role played by the mass $A = 132$ in asymmetric fission (cf. subsect. 4.2, 1); they further show, in the case p.l., that a considerable amount of energy, about 13 MeV, has been transformed into internal energy in the $A \sim 132$ fragment. And they show still further, in the case p.h., that the same amount of energy has also been transformed into internal energy, this time within the light fragment. Calling «source» of the polar $\alpha$-emission the fragment in which this amount of energy has been transformed, we may describe the situation as follows. One of the main sources of polar alphas emitted in the direction of the light fragment is a heavy fragment of mass about 132, and the value of the energy transformed in this source into internal energy is not too different from the energy that can be supplied by a giant dipole resonance. But other sources also contribute to the polar $\alpha$-emission; their mean mass is centred at $A \sim 141$.

The direction of emission of the polar alphas is that of the electrostatic $E$ field of the proton shells vibrating in the direction of the fission axis, whereas the direction of the emission of the equatorial alphas corresponds to the electromagnetic $E$ field of these vibrating proton shells.

In conclusion, the study of the emission of ternary alphas in the spontaneous fission of $^{252}$Cf by the Darmstadt-Heidelberg group [2, 15] furnishes new crucial arguments in favour of the role of spherical clusters such as $^{132}$Sn in an early stage of fission [19]. It suggests that collective pick-up reactions, occurring within a primordial dicleuster configuration of systems fissioning asymmetrically, and involving a great number of nucleons, can create giant resonance situations both in the heavy remnant of these reactions and in the light cluster which captures these nucleons. One of these kinds of repercussions of the initial dynamics of asymmetric fission could be the appearance of electric giant monopole resonances, which can explain the so-called non-statistical $\gamma$-ray component, reported by the Darmstadt-Heidelberg group [14].

REFERENCES