On the ³⁴Si emission of ²⁴²Cm

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Received: 18 October 2000 Communicated by V.V. Anisovich

Abstract. A ³⁴Si-decay of ²⁴²Cm has been reported by Oglobin *et al.* But an interpretation of this ³⁴Si emission as a ternary high-energy particle emission accompanying the fission of ²⁴²Cm can be proposed as well.

PACS. 23.70.+j Cluster decay – 25.85 Fission reactions

1 Introduction

At the 1998 International Nuclear Physics Conference, A. A. Oglobin *et al.* reported the preliminary observation of a ³⁴Si emission of the spontaneously fissioning nucleus ²⁴²Cm [1]. They found three events, releasing an energy of 69 (6) MeV, 76 (7) and 76 (7) MeV, which could be attributed neither to a contamination of the source by Si, nor to a cluster-activity of ²³⁸Pu, daughter of ²⁴²Cm, and calculations seemed to indicate that a Si emission due to the ternary fission of ²⁴²Cm was excluded. Thus the observations could be interpreted as a cluster-decay of ²⁴²Cm, with a partial half-life of 2×10^{23} s.

Recently , Oglobin *et al.* reported the observation of 15 events of ³⁴Si-decay of ²⁴²Cm, measured using trackrecording phosphate glass detectors [2]. They found a mean kinetic energy of $(81.0 \pm 1.9) \pm 2$ MeV, which is consistent with the expected energy of 82.97 MeV calculated from the clusterization energy of ²⁴²Cm in its ground state; 1.9 MeV is the mean-squared error for the experimental events, while 2.0 MeV is the systematical error in evaluating the range defect in glass. The registration threshold was about 69 MeV, due to the combined effect of absorbers and of the range deficit in the glass detector.

The aim of the present paper is to show that this ³⁴Siemission can be something else than a cluster decay. We first recall that there exist two different modes of light charged particle emission in ternary fission (sect.2). Then we show that the ³⁴Si emission cannot be explained as a low-energy equatorial mode, resulting from a stimulation by the double giant dipole resonance (sect. 3). But this emission can be explained as a high-energy isotropic mode: A ³⁴Si particle can be ejected from the fission fragment ¹⁶⁶Gd with an energy of 79.1 MeV in a collision with its partner ⁷⁶Ge, as a consequence of the tendency of nuclear matter to clusterize, even in a fission fragment, ¹⁶⁶Gd clusterizing according to (sect. 4)

$$^{166}\text{Gd} \to {}^{132}\text{Sn} + {}^{34}\text{Si.}$$
 (1)

THE EUROPEAN

 PHYSICAL JOURNAL A
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2 The two modes of light-charged-particle emission in ternary fission

We reported in 1996 [3] that the equatorial α -particle emission of ternary fission can be explained as resulting from the stimulation, by the double giant-dipole resonance (DGDR), of the potential α -radioactivity of the fission fragments. The Q_{α} -values of fission fragments are either negative, or extremely small, but the missing energy, *i.e.* the difference between, on the one hand, the effective mean Q_{α} , $\overline{Q_{\alpha}}_{\text{eff}}$, corresponding to the mean kinetic energy $\overline{E_{\alpha}} = 15.9$ MeV, according to [4]) of the ternary α -particles effectively emitted in the fission of actinide nuclei as different as ²³³U + n_{th} and ²⁵²Cf and, on the other hand, the mean value of the natural Q_{α} 's, $\overline{Q_{\alpha}}_{\text{nat}}$, of all the fission fragments is precisely equal to the energy of the DGDR. This situation may be written [5] as

Missing energy
$$= \overline{Q_{\alpha}}_{eff} - \overline{Q_{\alpha}}_{nat} = E_{DGDR}$$
. (2)

In the heavy nucleus 242 Cm, the GDR energy is equal to $(31.2A^{-1/3} + 20.6A^{-1/6})$ MeV, *i.e.* to 13.26 MeV, according to ref. [6], and the DGDR energy is approximately twice the GDR energy, according to ref. [7], *i.e.* equal to 26.52 MeV.

In the DGDR-stimulated light-charged-particle (LCP) emission, the Q_{α} 's of eq. (2) are replaced by the Q_{particle} -values, *e.g.* by the $Q_{1^{0}\text{Be}}$'s [8].

In this *equatorial* low-energy LCP emission, the particles are expelled at an angle of about 90 degrees from the fission axis. In the core-cluster collision leading to the rearrangement reactions of the fission process, a fierce transfer

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of $N'_{\rm p}$ protons from the core to the cluster occurs, with important consequences for both nascent fragments, in particular the appearance of a giant-dipole resonance due to the motion of proton phase against neutron phase. Only the two-phonon resonance has enough energy for stimulating the LCP radioactivity in such a way that it becomes observable.

Beside this equatorial low-energy LCP emission process, there exists another one, which is characterized by a higher energy and an isotropic distribution [5]. This phenomenon was observed by Chen *et al.* in fusion-fission reactions [9]. In the α -particle emission, the equatorial mode is so intense that the observation of the higher-energy mode is difficult. The weak emission, with a mean kinetic energy of about 25 MeV, observed by Heeg [10] at small angle from the fission axis, and for this reason called polar emission, is probably the high-energy isotropic mode [11]. We have suggested that this second mode of LCP emission could result from the ejection, in an internal collision within a binary system of fission fragments, of a cluster formed, in one of the fragments, from the valence nucleons of its doubly magic core [5].

This mechanism can be considered as a generalization of that proposed for explaining the second mode of fission [12,13]. For explaining the high neutron yield discovered in the Ba-Mo binary mass splits of 252 Cf, we have suggested [14] that carbon clusters can be formed in barium fission fragments from the valence shells of their 132 Sn doubly magic core, and presented arguments in favour of the existence of a dicluster system 132 Sn- 14 C in 146 Ba. Among these arguments, let us mention firstly the fact that the formation energy Q_1 of a carbon cluster in 146 Ba is greater than the formation energies of other carbon clusters in other barium isotopes, and secondly the great yield of the 138 Ba- 104 Mo binary mass split, showing that in 146 Ba- 106 Mo both partners contribute to the destruction reaction releasing 10 neutrons.

Whereas the ¹⁴C is destroyed in a collision of the two partners of the ¹⁴⁶Ba-¹⁰⁶Mo mass split, it is conceivable that less fragile clusters, and clusters which are better bound to the ¹³²Sn core than is the case for ¹⁴C could have a different behaviour. Consideration of the energy released in the clusterization of tellurium and xenon isotopes clearly shows that in the case of the helium clusters the emission of ⁴He in the fission of ²⁵²Cf must be favoured, and that in the case of the beryllium clusters the emission of ¹⁰Be must be favoured [5]. Consideration of the energy released in the clusterization of gadolinium isotopes clearly shows that in the case of silicium clusters the emission of ³⁴Si ($Q_1 = 42.15$ MeV) in the fissioning ²⁴³Am^{*} nucleus, formed by neutron capture in ²⁴²Am nucleus, could be favoured, if the great energy of the rearrangementreaction leading to the ¹⁶⁶Gd-⁷⁷Ga mass split ($Q_2 = 98.48$ MeV) is taken into account [5].

The hypothesis of the existence of a second mode of LCP emission in nuclear fission allows a prediction concerning the mass of the heaviest LCP which can be expelled in fission. Let us assume that the mass number of the heaviest cluster-emitting fission fragment which can be produced with a measurable yield is A_{lim} , then the heaviest observable cluster will have the mass number [5, 15]:

$$A_{\rm LCP} (\lim) = A_{\rm lim} - 132.$$
 (3)

3 The DGDR energy of ^{242}Cm is too small for ejecting a 81 MeV ^{34}Si cluster

Let us show that the 34 Si particles observed by Oglobin *et al.* with a mean kinetic energy of 81 ± 3.4 MeV cannot be the result of a DGDR-stimulated radioactivity.

Indeed, all natural $Q_{^{34}Si}$ -values of fission fragments are extremely small.

For example, in the mass splits 132 Sn- 110 Pd and 166 Gd- 76 Ge of 242 Cm, these Q-values are, for the fragments 132 Sn, 110 Pd and 76 Ge, respectively, equal to -11.79 MeV, + 4.82 MeV, and -16.27 MeV. Only 166 Gd has an exceptionally great value of + 42.15 MeV, due to the doubly magic character of the daughter 132 Sn of the clusterizing 166 Gd.

$$^{166}\text{Gd} \rightarrow ^{132}\text{Sn} + ^{34}\text{Si} + 42.15 \,\text{MeV}.$$
 (4)

But even in this case the DGDR-energy of 242 Cm, namely 26.52 MeV, cannot compensate the missing energy, since the missing energy is equal to

$$Q_{^{34}\text{Si eff}} - Q_{^{34}\text{Si nat}} = (166/132)81 - 42.15 = 59.71 \,\text{MeV}.$$
(5)

4 Estimation of the kinetic energy of ³⁴Si particles emitted according to the second mode

 $^{242}\mathrm{Cm}$ clusterizes according to

$$^{242}\mathrm{Cm} \rightarrow ^{208}\mathrm{Pb} + ^{34}\mathrm{Si}$$
 (6)

with an energy release $Q_1 = 96.52$ MeV. The energy released in the rearrangement reaction

$$^{208}\text{Pb} + {}^{34}\text{Si} \to {}^{166}\text{Gd} + {}^{76}\text{Ge}$$
 (7)

is equal to 85.92 MeV. Thus the energy stored in the 166 Gd- 76 Ge system can be as great as 182.44 MeV, and this energy can be shared between the two partners. And the maximal energy of the 166 Gd fragment should be equal to (76/242)182.44 = 57.29 MeV. However, the clusterization of 166 Gd according to eq. (4) releases an extra-energy of 42.15 MeV. In the new configuration of 166 Gd, 166 Gd*, the 34 Si-cluster could receive a kinetic energy equal to (132/166)(57.29 + 42.15) = 79.08 MeV.

Let us now assume that 166 Gd^{*} and 76 Ge make a collision. This collision should be essentially a collision between the 132 Sn core and the 76 Ge fragment, due to the elongated form of the 132 Sn- 34 Si, and the 34 Si cluster could escape with its own energy of 79.08 MeV. This energy is very close to that reported by Oglobin *et al.*

Table 1. Prediction of the KE of high-energy ternary Si particles emitted by 242 Cm according to the second LCP emission mode. The binding energies are from [19].

A(Gd fragment)	163	164	165	166	167	168
A(Si particle)	31	32	33	34	35	36
$Q_{\rm Si}$ (MeV)	38.13	40.93	40.61	42.15	40.32	40.94
A(Ge fragment)	79	78	77	76	75	74
Q_2 (MeV)	89.21	89.91	85.99	85.92	80.79	79.79
$\overline{Q}_{\rm tot}$ (MeV)	185.73	186.43	182.51	182.44	177.31	176.31
$KE(^{A}Gd)$ (MeV)	60.63	60.09	58.07	57.29	54.95	53.91
$KE(^{A}Gd^{*})$ (MeV)	98.76	101.02	98.68	99.44	95.27	94.85
$KE(^{A}Ge)$ (MeV)	125.10	126.34	124.44	125.14	122.36	122.40
$KE(^{A}Si)$ (MeV)	79.98	81.31	78.95	79.08	75.30	74.53

5 Discussion

The kinetic energy distribution of the high-energy particles has been carefully determined by Oglobin *et al.* [2]. It is centered at 81 ± 3.9 MeV. Low-energy particles, in particular equatorial low-energy ternary particles, could not be observed, due to the registration threshold at 69 MeV. A first question is: Are the observed particles the result of a cluster-decay, or are they high-energy ternary particles? But a second question is: Have the mass number of these particles and their atomic number been sufficiently well determined ?

In other words, is the competition between ${}^{34}Si$ decay and ${}^{34}Si$ ternary emission taken into consideration in Sects. 3 and 4, *i.e.* the intriguing competition, within one and the same nucleus ${}^{242}Cm$, between the emission of the primordial cluster ${}^{34}Si$ from the ${}^{242}Cm$ nucleus itself and the emission of a secondary cluster ${}^{34}Si$ from a particular fragment of this nucleus, the essential question we have to resolve ? Is it not necessary to ask first: Are not ternary particles other than ${}^{34}Si$ in competition with the ${}^{34}Si$ primordial cluster ?

The data on Si particles emitted in ternary fission taken into consideration by Oglobin *et al.* in their discussion [1,2] are those of ref. [17]. These data concern the fission of ²⁴³Am^{*}. But more recent data on ²⁴³Am^{*} exist: In their work at Lohengrin, published in 1996, Hesse *et al.* [18] report how they succeeded in observing not only ³⁴Si, but also ³²Si, ³³Si and ³⁵Si. Due to its greater yield, ³⁴Si was the first to be observed. In our opinion, the statement of [2] that ternary Si-particles formed in ²⁴³Am^{*} must have an energy smaller than 81 MeV has to be reconsidered, because it was based on the data of ref. [17].

In the following, we first show that Si particles with A values between 32 and 35 could be formed in the fission of 242 Cm, as a result of the second LCP emission mode, with energies close to the reported energy of 81 MeV (sect. 5.1), and we compare these predictions with similar predictions concerning the formation of P particles (sect. 5.2). We then compare all these predictions with the calculation of the energies of Si particles which could be formed in the ternary fission of 243 Am^{*} as a result of the second LCP

emission mode. These energies could be those of the Si particles effectively observed at Lohengrin (sect. 5.3).

5.1 Prediction of the kinetic energy of ternary Si particles emitted by ²⁴²Cm

In Table 1, we extend the discussion of sect. 4 to the formation of Si particles having A values differing from 34.

There, we first report the calculated value of the energy, $Q_{\rm Si}$, released in the formation of secondary Siclusters of given A in various gadolinium fragments from the valence nucleons of their ¹³²Sn core. It clearly appears that the Si particle having the greatest yield in the work of Hesse et al., ³⁴Si, is also the particle having the greatest clusterization energy $Q_{\rm Si}$, 42.15 MeV, and that the particle having the smallest yield, ³⁵Si, is also that having the smallest clusterization energy, 40.32 MeV. Table 1 further shows the value of the energy Q_2 released in the rearrangement reactions leading to these various gadolinium fragments, and the total energy released in the formation of these fragments, $Q_{tot} = Q_1 + Q_2$. But table 1 shows also how this energy is shared between Gd and Ge fragments as kinetic energy (KE) of the various heavy and light fragments. The KE of the heavy fragment, KE (^{A}Gd) , is changed into KE $(^{A}Gd^{*})$ by the phenomenon of clusterization, and KE $({}^{A}Gd^{*})$ is shared between Sicluster and ¹³²Sn core. Finally, the Si-cluster is expelled in the collision between Gd^{*}- and Ge- fragments with its own vibration energy, KE (A Si), reported in the last row.

It must be pointed out that the fission yield of the various Gd-fragments certainly plays an important role in the probability of formation of the secondary cluster, and information concerning this yield is seriously needed.

The most striking result furnished by Table 1 is the good agreement of the kinetic energies expected for $^{31-34}$ Si ternary particles with the KE distribution reported by Oglobin *et al.* in their fig. 4 [2]. However, more precisely, this KE distribution seems to be complex, with a main component at 79 MeV and a weaker component at 83 MeV, as shown by our own interpretation (fig. 1): the main component could correspond to ternary $^{33-34}$ Si, the weaker one, perhaps, to primordial 34 Si.

Table 2. Prediction of the KE of high-energy ternary P particles emitted by 242 Cm according to the second LCP emission mode.

A(Tb fragment)	164	165	166	167	168	169	170
A(P particle)	32	33	34	35	36	37	38
$Q_{\rm P}({\rm MeV})$	38.84	42.31	43.47	45.64	44.40	45.52	44.76
A(Ga fragment)	78	77	76	75	74	73	72
Q_2 (MeV)	84.02	84.80	82.19	82.58	78.80	78.08	73.19
$Q_{\rm tot}$ (MeV)	180.54	181.32	178.71	179.10	175.32	174.60	169.71
$KE(^{A}Tb)$ (MeV)	58.19	57.69	56.12	55.51	53.61	52.67	50.49
$KE(^{A}Tb^{*})$ (MeV)	97.03	100.00	99.59	101.15	98.01	98.19	95.25
$KE(^{A}Ga)$ (MeV)	122.35	123.63	122.59	123.60	121.71	121.93	119.22
$\mathrm{KE}(^{A}\mathrm{P})$ (MeV)	78.10	80.00	79.19	79.95	77.01	76.69	73.96

Table 3. Prediction of the KE of high-energy ternary Si particles emitted by ²⁴³Am^{*}.

A(Gd fragment)	163	164	165	166	167	168
A(Si particle)	31	32	33	34	35	36
$Q_{\rm Si}$ (MeV)	38.13	40.93	40.61	42.15	40.32	40.94
A(Ga fragment)	80	79	78	77	76	75
Q_2 (MeV)	98.69	100.44	98.34	98.48	95.04	94.73
$Q_{\rm tot}$ (MeV)	184.05	185.80	183.70	183.84	180.40	180.09
$KE(^{A}Gd)$ (MeV)	60.59	60.40	58.96	58.25	56.42	55.58
$KE(^{A}Gd^{*})$ (MeV)	98.72	101.33	99.58	100.40	96.74	96.52
$KE(^{A}Ga)$ (MeV)	123.46	125.39	127.73	125.58	123.98	124.50
$KE(^{A}Si)$ (MeV)	79.94	81.56	79.66	79.84	76.95	75.84



Fig. 1. Energy spectrum of the Si particles emitted from 242 Cm sources according to Oglobin *et al.* [2]. The dot-dashed curves are our own interpretation; they show a main component at 79 MeV and a smaller one at 83 MeV.

5.2 Prediction of the kinetic energy of ternary phosphorus particles emitted by $^{\rm 242}\rm{Cm}$

A contribution of particles with Z different from 14 has to be considered too. Let us report predictions concerning a contribution of phosphorus clusters. Table 2 demonstrates that a contribution of $^{33-35}P$ clusters formed in terbium fragments cannot be excluded on the sole basis of their kinetic energies, since ^{34}P , in particular, has an expected KE in perfect agreement with the main component of the KE distribution reported by Oglobin *et al.*

Alone the smaller yield of terbium fragments, as compared to the yield of gadolinium fragments — due to the odd Z value (Z = 65) — could justify a weaker contribution of P particles, as compared to Si particles.

5.3 Prediction of the KE of the Si particles emitted in the ternary fission of $^{\rm 243}{\rm Am}^*$

The work of Hesse *et al.* [18] does not furnish precise data on the KE of the Si particles emitted in the ternary fission of 243 Am^{*}. However, it is interesting to compare the relative fission yields of the $^{32-35}$ Si particles reported by these authors with energy calculations based on the hypothesis that these particles are formed according to the second LCP emission mode. Table 3 summarizes the calculations.

The striking result of these predictions is, firstly, that the KE of the ³²⁻³⁵Si clusters emitted by ²⁴³Am^{*} as highenergy ternary particles is very similar to that of the same silicium isotopes which could be emitted by ²⁴²Cm as highenergy ternary particles; secondly that the variation of the KEs as a function of A, for A = 33, 34 and 35 is perfectly similar to the variation of the fission yields of these particles as a function of A reported by Hesse *et al.* The different behaviour, for A = 32, could be due to a weaker fission yield of ¹⁶⁴Gd, as compared to that of ¹⁶⁶Gd.

This statement suggests that the primordial 34 Si of refs. [1,2] could be ternary Si clusters of various A value as well.

6 Conclusion

We have presented several arguments in favour of a possible contribution of ternary LCP emission to the phenomena presented as cluster-decay by the authors of refs. [1,2]. Among these arguments, the strongest one is the fact that ternary Si particles have been effectively observed at the Laue-Langevin Institute [18] and that we predict, for such ternary particles, exactly the same kinetic energy as that of the main component of the KE distribution reported by these authors. However, our predictions cannot explain the component of about 83 MeV apparently present in their KE distribution, and, furthermore, these predictions have still to be corrected for the variation of yield, as a function of mass number, of the Gd- and Tb-fragments occurring in the fission of ²⁴²Cm or ²⁴³Am^{*}. Thus, the reality of a competition between primordial and secondary cluster emission is still questionable, and further work is needed.

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