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Temperature of low-energy ternary fission from the dependence of particle yields on the mass of the fissioning system

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In thermal neutron-induced and spontaneous fission the addition of two neutrons to a neutron-even system produces only minor changes to the shape, kinetic energy, and temperature of the scission configuration. If these changes are assumed negligible and if ternary fission is associated with a statistical process then the ratio of ternary fission yields for systems differing by two atomic mass units can be used to infer a nuclear temperature. The yields of hydrogen, helium, lithium, and beryllium isotopes ejected perpendicular to the direction of the main fragments from ^{233,235}U($n_{\rm th}, f$), ^{239,241}Pu($n_{\rm th}, f$), and ^{250,252}Cf spontaneous fission give a nuclear temperature $T = 1.24 \pm 0.10$ MeV. The yield of polar α particles from ^{233,235}U($n_{\rm th}, f$) give $T = 1.13 \pm 0.24$ MeV. These results are in agreement with other inferred low-energy ternary fission temperatures and support the idea that both equatorial and polar ternary fission involve a statistical process where the ejected particles are in equilibrium with a heat bath with a temperature slightly hotter than 1 MeV.

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I. INTRODUCTION

An understanding of the nature of ternary nuclear fission has not yet been obtained despite extensive attempts over the past 50 years. The high-energy cost to produce ternary-fission light-charged particles (LCP) clearly rules out standard particle evaporation from nonaccelerating hot nuclear matter [1,2]. Many have used this fact to rule out an evaporation process, instead of considering how an evaporation process might be altered in the presence of a rapidly changing hot nuclear fluid. Many dynamical models of ternary fission exist and have been reviewed in Ref. [2]. These include an extension of the theory of particle emission from actinide ground states to a rapidly evolving system in the last phase of the fission process [3], and models based on the assumption that ternary-fission LCP are formed as a result of two random neck ruptures during the time interval of one single-particle period [4]. More recently, some fraction of spontaneous ternary-fission events have been viewed as a cold quantum mechanical rearrangement of nucleons from the ground state of an initial nucleus to the ground or low-excited states of three final fragments [5].

A statistical model of ternary fission was developed by Fong [6,7]. In this model the fissioning system is assumed to remain in statistical equilibrium and thus the initial conditions for ternary fission events are governed by phase-space arguments. These initial conditions, in conjunction with trajectory calculations, give α-particle energy and angular distributions that are consistent with experimental results [6]. Despite this apparent success, a statistical process has been dismissed because the high-energy cost for generating ternary-fission LCP is inconsistent with the observed ternary-fission probabilities within the framework of a purely statistical model [2]. However, an apparent nuclear temperature associated with low-energy ternary fission has been obtained using isotope thermometry [8]. This method utilizes double isotope yield ratios from the same fissioning system to infer a nuclear temperature. Ternary-fission yield data from fissioning systems from ^{229}Th to ^{252}Cf were used. The inferred nuclear temperature of low-energy ternary fission is $T = 1.10 \pm 0.15$ MeV. Isotope thermometry is based on the general assumption that thermal equilibrium is achieved. The inferred ternary fission temperature of $\sim$1 MeV could only be viewed as fortuitous if ternary fission is a cold quantum mechanical and/or purely dynamical process.

A combined statistical and dynamical model of ternary fission has been proposed [9]. In this model potential ternary-fission particles are first produced beyond the surface of scission configurations via a statistical process and then ejected dynamically by the sudden rearrangement following scission. Key features of the combined statistical and dynamical model of ternary fission are that the initial location and velocity of LCP are determined by phase-space arguments and the statistical particle generation mechanism does not need to produce the full energy required for ternary fission.

To confirm (or otherwise) the importance of a hot statistical process in the emission of ternary fission LCP, we present a new method for inferring a nuclear temperature associated with low-energy ternary fission using the yield of LCP as a function of the mass of the fission system in thermal neutron induced and spontaneous fission. This method is independent of isotope thermometry [8], which uses the dependence of ternary fission yields as a function of LCP isotope within a fixed fissioning system.

II. THEORY

The thermal neutron induced or spontaneous fission of two neutron-even isotopes differing by two neutrons is very similar. Examples of such pairs of isotopes with similar fission properties are ^{234,236}U, ^{240,242}Pu; and ^{250,252}Cf. For each of these pairs of isotopes the potential energy surfaces are similar, and thus the temperatures, shapes, and collective kinetic energies at scission will be similar. If these changes are assumed negligible, and if ternary fission is associated with a statistical process, then the ratio of ternary-fission yields for a pair of neutron-even isotopes differing by two neutrons will be controlled by phase-space arguments and given by the following expression:

$$Y(A_f+2)/Y(A_f) = \exp(-\Delta B_E/T), \qquad (1)$$

where A_f is the mass number of the lighter of the isotope pair, T is the nuclear temperature, and ΔB_E is the change (difference) in the LCP binding energy between the isotope pair as follows:

$$\Delta B_E = B_E(A_f+2) - B_E(A_f). \qquad (2)$$

The particle binding energy at scission can be expressed as follows:

$$B_E = M_D + M_{\rm LCP} - M_P + B_E^{\rm Shift}, \qquad (3)$$

where $M_{\rm LCP}$ is the ground-state mass of the LCP and M_D and M_P are the modified-liquid-drop-model [10] masses of the daughter and parent scission configurations assuming the same shape for both the parent and daughter systems. $B_E^{\rm Shift}$ corrects for the change in the distance between mass centers of the two main nascent fragments, at scission, produced by the generation of the ternary fission LCP. $B_E^{\rm Shift}$ can be estimated assuming that the center of mass of each of the halves of the scission configuration remains fixed during the generation of the LCP either near the neck region in the case of equatorial ternary fission or near the polar regions in the case of polar ternary fission. For equatorial ternary fission $B_E^{\rm Shift}$ can be expressed as follows:

$$B_E^{\rm Shift} = 0.36({\rm MeV\,fm})\frac{Z_D^2}{r_S}\left\{\frac{A_D}{A_P} - \left(\frac{A_P}{A_D}\right)^{1/3}\right\}, \qquad (4)$$

where r_S is the distance between mass centers of the nascent fragments of the parent and Z_D, A_D, and A_P are the atomic and mass numbers for the daughter and parent systems. For polar ternary fission $B_E^{\rm Shift}$ is given by the following:

$$B_E^{\rm Shift} = 0.36({\rm MeV\,fm})\frac{Z_D^2}{r_S}\left\{\frac{r_S}{r_S-\Delta_S} - \left(\frac{A_P}{A_D}\right)^{1/3}\right\}, \qquad (5)$$

where the shift in the nascent fragment producing the polar LCP is as follows:

$$\Delta_S \sim 2.45({\rm fm})\frac{A_{\rm LCP}}{A_P}\left[(A_P/2 - A_{\rm LCP})^{1/3} + A_{\rm LCP}^{1/3}\right]. \qquad (6)$$

The equatorial ternary fission particle binding energies for ^{240}Pu and ^{242}Pu and the difference between these binding energies (ΔB_E) are given in Table I. The distance between mass centres at scission is assumed to be 2.6 times the nuclear radius of the corresponding spherical system [9]. Even though the individual particle binding energies depend on the compound system mass and charge, and strongly on the assumed elongation at scission, the change in the particle binding energies is very insensitive to these quantities. Changing the Z of the isotope pair from 92 to 98, and dramatic charges in the assumed shape of the emitting system, produce typical changes in ΔB_E of less than a few hundredths of a mega-electron-volt.

The particle binding energy changes (ΔB_E) decrease with increasing mass of the LCP with fixed $Z_{\rm LCP}$. This behavior is easily explained. Heavy nuclei near the valley of stability have a mass-to-charge ratio $A/Z \sim 2.6$. For LCP with $A/Z < 2.6$ (e.g., an α particle) the addition of two neutrons to the fissioning system makes emission more difficult and this is reflected in the positive values of ΔB_E for these particles. The reverse is true for LCP with $A/Z > 2.6$ (e.g., ^{8}He). Therefore, if ternary fission is associated with a statistical process then the yield ratios $Y(A_f+2)/Y(A_f)$ should be very insensitive to both A_f and Z_f and increase with increasing mass of the LCP with fixed $Z_{\rm LCP}$. If this is the case then the ternary fission yield ratios, $Y(A_f+2)/Y(A_f)$, could be used to infer a nuclear temperature via Eq. (1).

TABLE I. Equatorial ternary fission (ETF) particle binding energies for ^{240}Pu and ^{242}Pu, and the difference between these binding energies (ΔB_E).

Isotope	$B_E^{\rm Scission}$ (ETF) (MeV)		ΔB_E (ETF) (MeV)
	^{240}Pu	^{242}Pu	
^{1}H	7.465	7.848	+0.383
^{2}H	9.953	10.116	+0.163
^{3}H	8.520	8.464	−0.056
^{4}He	−3.749	−3.420	+0.329
^{6}He	4.930	4.813	−0.117
^{8}He	12.918	12.346	−0.572
^{7}Li	2.576	2.849	+0.273
^{8}Li	5.371	5.420	+0.049
^{9}Li	6.255	6.075	−0.180
^{9}Be	−3.645	−3.195	+0.450
^{10}Be	−5.687	−5.473	+0.214
^{11}Be	−1.301	−1.317	−0.016
^{12}Be	0.538	0.291	−0.247

III. RESULTS

Equatorial ternary fission yield ratios, $Y(A_f+2)/Y(A_f)$, are shown in Fig. 1 for ^{233,235}U$(n_{\rm th}, f)$, ^{239,241}Pu$(n_{\rm th}, f)$, and ^{250,252}Cf spontaneous fission. The horizontal axis $[A_{\rm LCP} + 2(Z_{\rm LCP}-1)]$ was chosen to conveniently separate the hydrogen, helium, lithium, and beryllium data. Notice the increasing trend in the yield ratio with increasing mass of the LCP with fixed $Z_{\rm LCP}$. The helium data yield ratios are insensitive to A_f and Z_f. The nonhelium data are not as conclusive. To infer a nuclear temperature associated with low-energy ternary fission, Eq. (1) was used in conjunction with the ΔB_E given in Table I. The nuclear temperature was adjusted to minimize the chi-squared fit of the model calculations to all data shown in Fig. 1. The inferred temperature for low-energy ternary fission is $T = 1.24 \pm 0.10$ MeV. The solid curves in Fig. 1 show the best fit with $T = 1.24$ MeV. Three of the 25 yield ratios shown in Fig. 1 differ by more than 3-σ from the solid curves. These are the deuteron yield ratio for the isotope pair ^{239,241}Pu$(n_{\rm th}, f)$ and the ^{9}Li and ^{10}Be data for the ^{233,235}U$(n_{\rm th}, f)$ pair.

As stated under Sec. II, the binding energy changes (ΔB_E) are very insensitive to the assumed shape of the parent/daughter systems. If the binding energy shifts (corrections) given by Eq. (4) are set to zero then the extracted ternary fission temperature increases by only 5% to 1.30 ± 0.10 MeV.

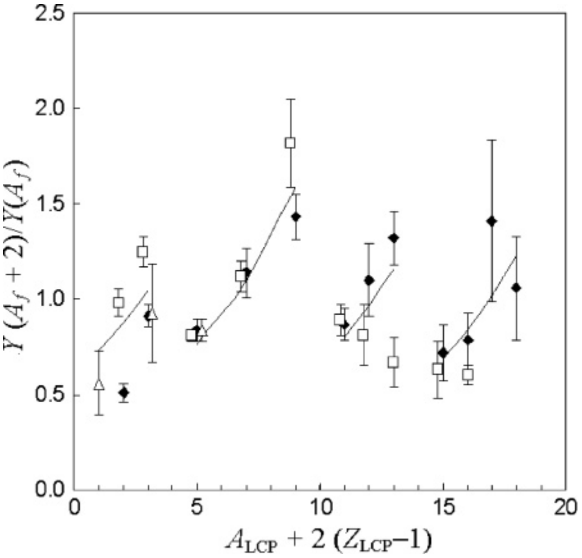

FIG. 1. The ^{233,235}U(n_{th}, f) (open squares); ^{239,241}Pu(n_{th}, f) (solid diamonds); and ^{250,252}Cf (open triangles) spontaneous fission equatorial ternary fission yield ratios, $Y(A_f+2)/Y(A_f)$, versus $A_{LCP}+2(Z_{LCP}-1)$. These yield ratios were determined by a combination of various experimental results [11–18]. The solid curves show a model calculation (see text).

If the parent/daughter systems are assumed to be spherical liquid drop model (LDM) nuclei or two well-separated LDM symmetric fission fragments, then the trends in the yield ratios shown in Fig. 1 are still well reproduced but with temperatures of 1.39 ± 0.11 and 1.34 ± 0.11 MeV, respectively. Assuming emission occurs from a single spherical system or from well-separated spherical fragments is an extreme unrealistic case. These results show that the extracted equatorial ternary fission temperature of $T = 1.24 \pm 0.10$ MeV is very insensitive to the details of the statistical emission process.

Polar ternary fission is a much rarer process than equatorial ternary fission, and thus the polar ternary fission data set is much more limited. An α-particle polar ternary fission yield ratio of 0.78 ± 0.04 has been measured for the ^{233,235}U(n_{th}, f) reactions [19]. The corresponding ΔB_E is 0.280 MeV, and thus the inferred temperature (for polar emission) is $T = 1.13 \pm 0.24$ MeV. This extracted temperature increases by 5% if the binding energy shift given by Eq. (5) is set to zero and by less than 15% if the emission is assumed to be from a spherical LDM nucleus or from two-well separated LDM symmetric fission fragments.

IV. DISCUSSION

In the present study, it is assumed that there is a universal temperature for all low-energy ternary fission reactions independent of reaction type and mass and charge of both the LCP and the fissioning system. There must, of course, be small temperature variations with all of these quantities. However, to correct for any possible dependence of the ternary fission temperature on reaction type, and the mass and charge of

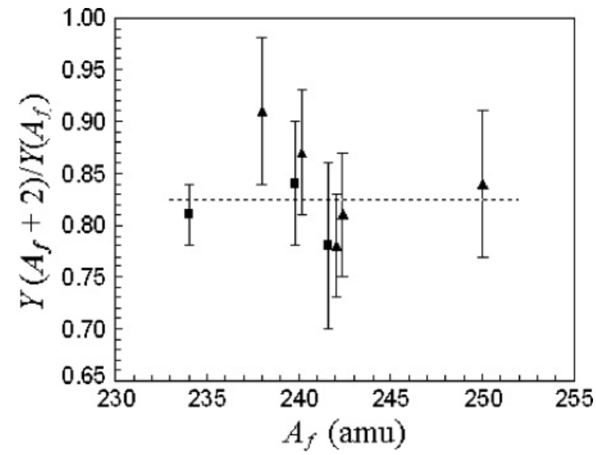

FIG. 2. Measured α-particle yield ratios versus A_f for spontaneous fission (triangles) and thermal-neutron-induced fission reactions (squares). These yield ratios were determined by a combination of various experimental results [2,15–18,20]. Some of the points have been shifted by not more than 0.4 atomic mass units to avoid clutter. The horizontal line shows the uncertainty weighted average at a yield ratio of 0.825.

the LCP and/or fissioning system, would require a detailed knowledge of the emission mechanisms.

It is generally believed that the dependence of the ternary-fission ^{4}He emission probability as a function of the mass of the fissioning system is governed by the α-cluster preformation probability S_α [20]. The simple model calculations presented in this article provide an alternative description of the mass dependence of ternary fission ^{4}He emission. The simple statistical model presented here also reproduces the dependence of the ^{6}He and ^{8}He emission on the mass of the fission system. Based on the present work the α-particle yield ratio $Y(A_f+2)/Y(A_f)$ should be insensitive to the A and Z of the fission system and have a value given by Eq. (1). Figure 2 shows measured α-particle yield ratios for both spontaneous fission and thermal neutron induced reactions.

The measured α-particle yield ratios scatter about 0.825, not the value of 0.77 predicted by Eq. (1) with $T = 1.24$ MeV. This discrepancy of $\sim$7% could be because of the assumption of a universal temperature for all reactions and for all LCP and/or the assumption that the addition of two neutrons does not change the shape or collective energy at scission. A drop in scission temperature of ΔT caused by the additional of two neutrons can be corrected for using the expression

$$Y(A_f+2)/Y(A_f) = \exp\left(-\frac{\Delta B_E}{T}\right)\left(1-\frac{\Delta T\ \bar{B}_E}{T^2}\right), \qquad (7)$$

where $\bar{B}_E$ is the average particle binding energy for the isotope pair. A value of $\Delta T/T \sim 0.01$ will increase the α-particle yield ratio by $\sim$3%. The temperature T in Eq. (7) should also vary with the LCP. This is because the rarer particles require additional energy to be emitted. This depletes the excitation energy of the parent system and lowers the effective temperature for the rarer emissions. The temperature for dominant ^{4}He emission will therefore be larger than a

TABLE II. Summary of evidence for a temperature of ∼1 MeV for scission configurations in low-energy fission.

Basis for the inferred temperature		Inferred temperature (MeV)
Mean kinetic energy of fragments	Dynamical calculations like those performed in Ref. [9]	1.1
Isotope thermometry	Ref. [8]	1.10 ± 0.15
^{10}Be 3.368 MeV γ-ray	Ref. [23]	1.0 ± 0.1
ETF yield ratios	Present work	1.24 ± 0.10
α-PTF yield ratio	Present work	1.13 ± 0.24

temperature averaged over all light charged particles, whereas the rarer particles should have an effective temperature that is lower. Correcting for this effect would require a detailed knowledge of the emission mechanism.

Assessing any effects of small changes in the shape and collective energy at scission on the yield ratios would require a detailed multidimensional model of the passage of fissioning systems to their scission configurations and a detailed model of the emission mechanism. This is beyond the scope of the present study.

It must be emphasized that the aim of the present article is to show the importance of a statistical process to the emission of light-charged particles in low-energy ternary fission; without making detailed assumptions about the emission mechanism, expect that standard particle evaporation can be ruled out as mentioned in the introduction. The results presented here cannot be used to distinguish among different models containing a statistical process involving a nuclear temperature of ∼1 MeV and do not prove the correctness of the detailed assumptions made in the combined statistical and dynamical model of ternary fission [9]. The present work does not rule out the possibility that quantum-mechanical ejection processes, like those discussed in Refs. [5,21,22], play a central role in ternary fission, because thermal fluctuations can be incorporated into these quantum mechanical emission process.

V. CONCLUSIONS

The ^{4,6,8}He ternary-fission yield ratios $Y(A_f+2)/Y(A_f)$ are insensitive to the A and Z of the fissioning system and increase in increasing mass of the helium. This behavior is easily understood if a statistical process plays an important role in ternary fission. Because of the larger uncertainties, clear conclusions cannot be drawn from the existing hydrogen, lithium, and beryllium data. The dependence of the equatorial ternary fission yield ratios on the mass of the light charged particles is consistent with a nuclear temperature of $T \sim 1.2$ MeV at the time of scission. The single yield ratio datum for polar ternary fission is consistent with this temperature.

The evidence for a temperature of ∼1 MeV at scission, in low-energy fission reactions, is summarized in Table II. Some of the reasons for believing the scission configurations in low-energy ternary fission have a temperature of ∼1 MeV, are weak when view individually. However, viewed as a whole, the evidence summarized in Table II strongly implies that scission configurations in ternary fission have a temperature of ∼1 MeV that is, at most, only weakly dependent on the mass and charge of the light particle. Based on the consistency of temperatures associated with low-energy ternary fission, obtained by a variety of different methods, it is concluded that ternary fission involves a statistical process where the ejected particles are in equilibrium with a heat bath with a temperature slightly hotter than 1 MeV.

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