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Fission and Quasi-Fission in Reactions with Deformed Nuclei

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Abstract—The mass-energy distributions of fragments formed in reactions ^{16,18}O + ^{232}Th, ^{238}U, and ^{22}Ne + ^{232}Th, ^{238}U at energies near the Coulomb barrier are measured to study the role of multimodal fission in the reactions of light ions with strongly deformed actinide nuclei. It is found that at these energies, multimodal fission affects significantly the mass–energy and angular distributions of fragments that results in an increase of the width of mass distributions and large angular anisotropy as in the case of quasifission.

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INTRODUCTION

In collisions between strongly deformed nuclei, their mutual orientation has a substantial impact on the reaction dynamics. Analysis of the mass–energy and angular distributions of fragments shows that at energies near and below the Coulomb barrier, a considerable contribution from the quasi-fission process to the capture cross section of the reaction is observed in reactions between $^{40,\,48}$Ca ions and strongly deformed isotopes $^{152,\,154}$Sm, while no quasi-fission is detected for reactions with spherical ^{144}Sm nuclei [1].

The dynamics of the reactions between such light ions as C, O, F, Ne, and actinide nuclei has been widely studied by many scientific groups. Abnormally high values obtained experimentally for the angular anisotropy of fragments [2] and an increase in the dispersion of mass distributions [3] in the sub-barrier region have been explained by manifestation of the quasi-fission process caused by orientation effects in reactions with deformed nuclei.

An abnormally high anisotropy of fragments at sub-barrier energies of interaction was detected for the first time in the reaction between ^{16}O and deformed ^{238}U nuclei. Cross sections of the formation of evaporative residue were also measured for this reaction at energies above and below the barrier [4]. Calculations using a statistical model with allowance for the deformation of ^{238}U reproduced the obtained experimental cross sections. The analysis of evaporative residues performed in [4] thus showed there was no suppression of the fusion channel in the sub-barrier region. And therefore, high angular anisotropy and an enhanced width of the mass distribution of fragments are not the reasons to expect the presence of the orientation-dependent quasi-fission for this system.

Considerably high experimental values of the angular anisotropy of asymmetrically divided fragments were observed in radiochemical measurements of the angular distributions of fission fragments in the 15 MeV proton-induced fission of Th [5]. It was found that the angular anisotropy of symmetric fission fragments was ~40% less than for asymmetric ones. The experimental values of angular anisotropy were in good agreement with the results from theoretical calculations performed by assuming the existence of two saddle points that led to symmetric and asymmetric mass division, respectively.

The transition state model traditionally used in theoretical analyses of the angular distributions of fission fragments reproduces experimental values of angular anisotropy fairly accurately for reactions with neutrons and light charged particles, whereas for reactions with more massive ions (e.g., C, O, and heavier), theoretical predictions are regularly much lower. The angular distributions of fission fragments obtained in reactions ^{16}O + ^{232}Th, ^{12}C + $^{235,\,236,\,238}$U were analyzed theoretically in [6] in a wide range of energies below and above the barrier. High values of angular anisotropy obtained experimentally for these reactions were described using a dynamic model for calculating the angular distributions of fission fragments that considered thermal fluctuations of the orientational degrees of freedom, while the evolution of the fission nucleus was considered to be a stochastic process. Moreover, the influence of the complete fusion reaction entrance channel properties on anisotropy of angular distributions at sub-barrier collision energies was taken into account for these systems.

Table 1. Properties of the systems studied in this work: A_{CN} is the atomic number of the compound nucleus, Z_1Z_2 is the Coulomb parameter, η_0 is the asymmetry of the entrance channel, E_{lab} is the energy of the incident particle, $E_{c.m.}/E_{Bass}$ is the energy relative to the interaction barrier, E^* is the initial excitation energy of the compound nucleus, and σ_M is the standard deviation of the experimental mass distribution

Reaction	A_{CN}	Z_1Z_2	η_0	E_{lab}, MeV	$E_{c.m.}/E_{Bass}$	E^*, MeV	σ_M, amu
$^{16}O + ^{232}Th$	^{248}Cf	720	0.87	84	0.97	42	17.0 ± 0.3
				96	1.10	53	18.2 ± 0.2
$^{18}O + ^{238}U$	^{256}Fm	736	0.86	85	0.96	40	19.3 ± 0.4
$^{16}O + ^{238}U$	^{254}Fm	736	0.87	89	1.01	45	18.9 ± 0.3
				101	1.14	56	19.0 ± 0.2
$^{22}Ne + ^{232}Th$	^{254}Fm	900	0.83	108	0.99	45	21.4 ± 0.3
				114	1.04	50	19.9 ± 0.2
				122	1.11	58	20.7 ± 0.2
$^{22}Ne + ^{238}U$	^{260}No			107	0.95	41	20.5 ± 0.4

The increase in the width of the mass distribution of fragments upon a drop in the energy in the sub-barrier region can be explained using the multi-modal concept of nuclear fission, which assumes that the mass–energy distributions of fragments are a superpositioning of the distributions of several independent fission modes with substantially different properties.

Mass–energy distributions of fragments of the reactions $^{16,18}O + ^{232}Th$, ^{238}U, and $^{22}Ne + ^{232}Th$, ^{238}U were measured at energies near the Coulomb barrier in order to clarify the question about the presence of the quasi-fission process in reactions of light ions with strongly deformed targets.

EXPERIMENTAL

Our experiments were performed at the Flerov Laboratory of Nuclear Reactions in Dubna, using the $^{16,18}O$ and ^{22}Ne ion beams, extracted from the U400 cyclotron, at energies near the Coulomb barrier. The energy resolution of the beams was ~2%. Targets were obtained by spraying $^{238}UF_4$, $^{232}ThF_4$ (120–200 μg cm^{-2}) on carbon films 40–50 μg cm^{-2} thick.

The products of binary reactions were registered using a CORSET double-arm time-of-flight spectrometer [7], each arm of which consists of start and position-sensitive stop detectors based on microchannel plates. The angular acceptance of each arm was ±10° in the plane of the reaction and ±8° outside it. The angular resolution of the spectrometer was 0.3°, and its time resolution was ~150 ps. The mass resolution of the spectrometer under these conditions was 2–3 amu.

The mass-energy distributions of primary binary fragments were measured using the double velocity method [7]. Data processing was based on the laws of momentum and mass number conservation by assuming that the mass of the composite system was equal to $M_{target} + M_{projectile}$. Neutron evaporation before scission was not taken into account. However, since the probability of the evaporation of more than three prescission neutrons in the studied reactions is very low even at the highest interaction energies, and the mass resolution of the spectrometer is 2–3 amu, neutron emission will not affect the mass-energy distributions. The energy losses of reaction products in the target and start detectors' foils were considered in processing the data. Analysis based on the kinematic diagram (the velocity vectors of registered reaction products) in the center-of-mass system allowed us to reliably separate the binary reaction channel from the products of sequential fission, incomplete fusion reactions, and reactions on impurity atoms in the target [7].

The characteristics of the studied reactions are shown in Table 1.

RESULTS AND DISCUSSION

The mass–energy distributions of binary fragments of the $^{22}Ne + ^{232}Th$ reaction are shown in Fig. 1. The upper panels show the mass yields of the fragments, normalized to 200%. The middle and lower panels show distributions of the average total kinetic energy (TKE) and its variance as a function of fragment mass. The lines show the calculations made using the liquid-drop model (LDM). The mass yield of fragments in the asymmetric region is higher than the predictions of the LDM for all of the studied energies. The same trend is observed in the dependence of the average TKE on the mass of the fragment. It should be noted that the deviation from the results of LDM calculations decreases with increasing the excitation energy of the compound nucleus. The mass–energy distributions of binary fragments of the reactions of $^{16,18}O$ with a ^{238}U target are shown in Fig. 2.

The hypothesis of coexistence of two independent (symmetric and asymmetric) fission modes was first proposed and applied successfully by Turkevich and

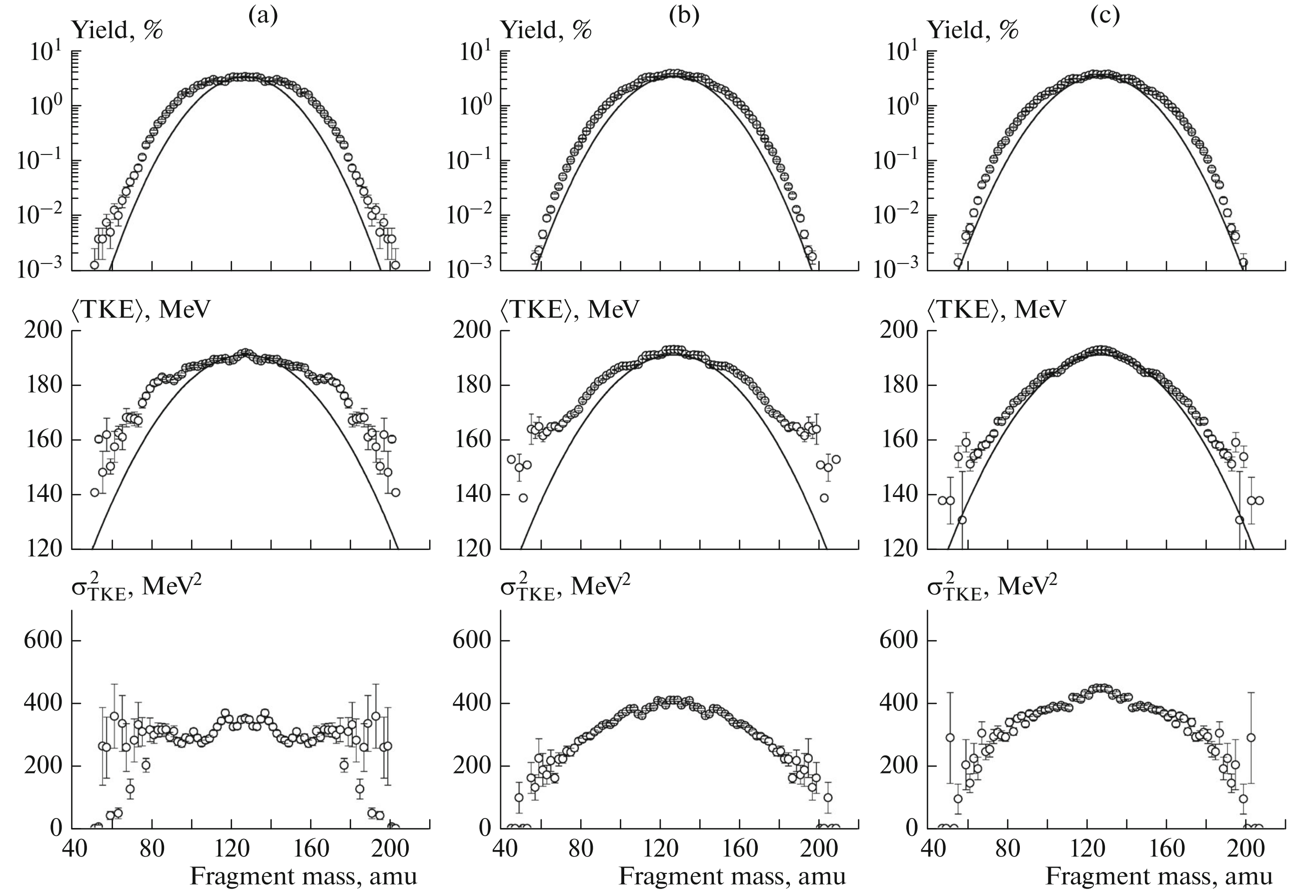

Fig. 1. Mass-energy distributions of ^{22}Ne + ^{232}Th reaction fragments at excitation energy of the compound nucleus $^{254}Fm^*$ of (a) 45, (b) 50, and (c) 58 MeV. From top to bottom: mass yields, average total kinetic energies ⟨TKE⟩ and their variance σ^2_{TKE} in dependence on the fragment mass; (solid lines) results of calculations based on the liquid-drop model.

Niday [8] to analyze the mass–energy distributions of fragments from fast neutron induced fission of ^{232}Th. Subsequent development of the multi-modal approach was stimulated by experimental studies in vicinities of Po and Fm nuclei, whose mass–energy distributions exhibited pronounced structures. Numerous theoretical studies showed that liquid-drop and shell effects in a fissioning nucleus lead to the emergence of several valleys on the potential energy surface that are responsible for the formation of mass–energy distributions of independent modes. The modern classification of fission modes usually uses the notation in [9], which predicted the existence of three modes: one symmetrical (*S*) and two asymmetric: Standard 1 (*S*1), which is due to the influence of proton $Z = 50$ and neutron $N = 82$ shells, and Standard 2 (*S*2), which is usually associated with deformed shells Z ≈ 54–56 and N ≈ 86–88. These ideas were developed further in [9, 10], where it was shown there can be more than three valleys. The Standard 3 (*S*3) mode associated with manifestation of neutron shell $N = 50$ was predicted in particular.

In the modal fission of actinide nuclei, the contribution from asymmetric fission modes depends on the nucleon composition and excitation energy of the fissioning nuclei. As the energy of excitation of a composite nucleus grows, the contribution from asymmetric modes diminishes due to the washing out of shell effects. The shell nature of modal fission leads to more compact configurations at scission, which in turn results in higher values of total kinetic energy, compared to classical fission. As can be seen from Fig. 2, there is an increased yield of fragments in the mass region of ~70–90 amu. The average TKE of these fragments is also higher than the LDM predictions. The properties of mass–energy distributions thus show features characteristic of modal fission.

Figure 3 shows the results from decomposing the mass distribution of fragments formed in the reaction ^{22}Ne + ^{232}Th at an excitation energy of 45 MeV. The mass distribution was described by Gaussians corresponding to four modes of fission (*S*, *S*1, *S*2, *S*3). The peak positions for asymmetric modes were fixed at the values obtained for the corresponding closed shells,

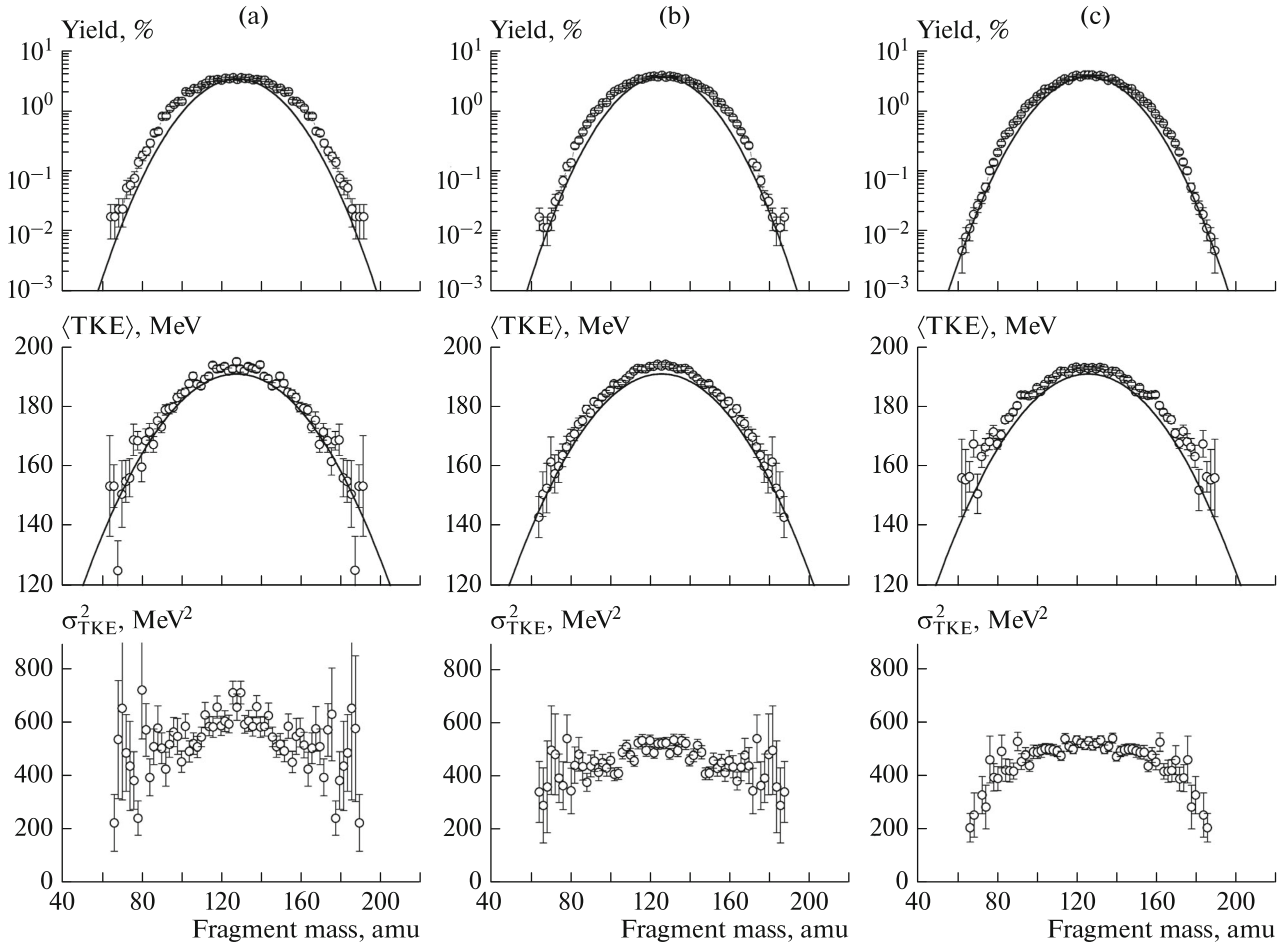

Fig. 2. Mass-energy distributions of fragments of reactions of ions (a) ^{16}O and (b, c) ^{18}O with ^{238}U target, leading to the formation of composite nuclei ^{254}Fm* and ^{256}Fm* at excitation energies of 40, 45, and 56 MeV. From top to bottom: mass yields; average total kinetic energies ⟨TKE⟩ and their variance, σ^2_{TKE} in dependence on the mass of the fragment; (solid lines) results from calculations based on the liquid-drop model.

based on the simple hypothesis of unchanged charge density.

The dependences of the width of mass distributions on the energy of interaction for all of the studied reactions are shown in Fig. 4. Standard deviations of the experimental mass distributions are indicated by symbols, while lines show the calculations of the LDM. As is seen from Fig. 4, when the energy is lower than the Bass barrier, the width of the mass distribution increases. Figure 4 also shows data for the reaction ^{18}O + ^{208}Pb, which was studied in detail in [11–14]. The results from a multicomponent analysis of the fragments showed that four different modes of fission (*S*, *S*1, *S*2, *S*3) were present in the mass–energy distributions up to excitation energies on the order of 80 MeV. Theoretical calculations of the prescission forms of fissioning nuclei $^{224,\ 226}$Th, along with the results from measuring the multiplicity of neutrons and gamma quanta, confirmed this conclusion. The presence of orientation-dependent quasi-fission was obviously not expected for this reaction, since both reaction partners (^{18}O, ^{208}Pb) were spherical nuclei, and all structural features in the mass–energy distributions were associated with the manifestation of modal fission. As is seen in Fig. 4, the dependence of the mass distribution variance for the reaction ^{18}O + ^{208}Pb shows the same behavior as for reactions on deformed targets. Thus, the enhanced values of the variance of mass distributions in the sub-barrier region in reactions of light ions with actinide target nuclei can be explained by the manifestation of asymmetric fission modes.

CONCLUSIONS

An increased yield of fragments in the mass region of ~70–90 amu was found for compound nuclei ^{248}Cf* and $^{254,\ 256}$Fm* formed in reactions of $^{16,\ 18}$O and ^{22}Ne

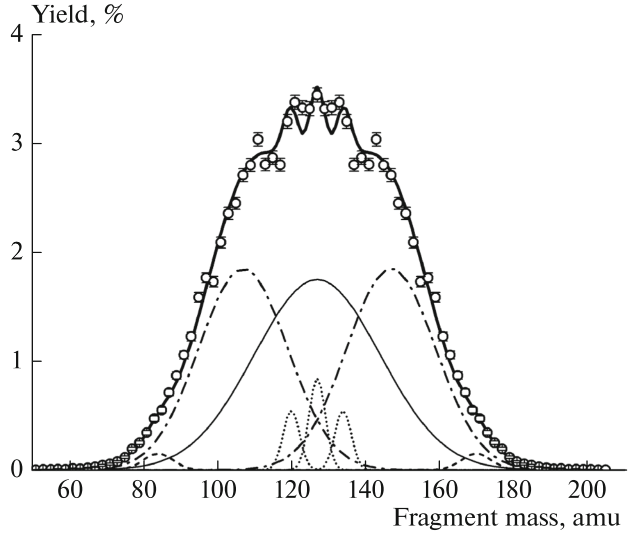

Fig. 3. Mass distribution of fission fragments formed in the reaction $^{22}Ne + ^{232}Th$ at an excitation energy of the compound nucleus $^{254}Fm^*$ of 45 MeV and the results from its decomposition into fission modes: (solid line) *S*, (dotted lines) *S*1, (dashed-and-dotted lines) *S*2, and (dashed lines) *S*3.

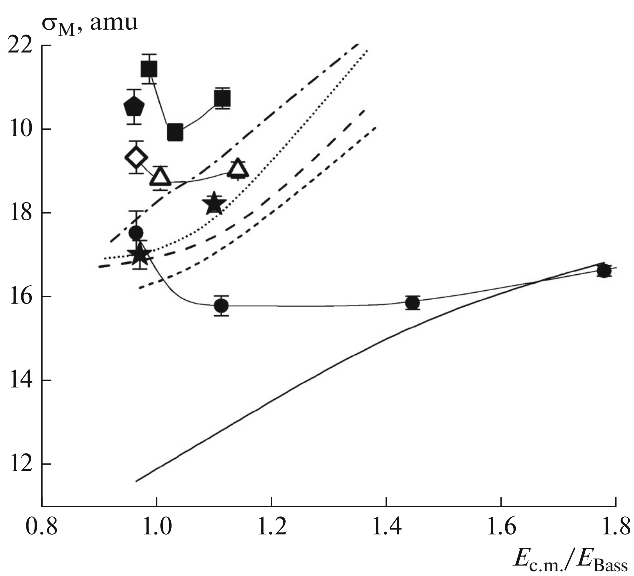

Fig. 4. Dependence of the width of the mass distribution of fragments on the energy introduced into the system relative to that of the Bass barrier for all of the studied reactions. Symbols show the experimental values of standard deviations of mass distributions: (squares) $^{22}Ne + ^{232}Th$, (pentagons) $^{22}Ne + ^{238}U$, (stars) $^{16}O+^{232}Th$, (triangles) $^{16}O + ^{238}U$, (diamonds) $^{18}O + ^{238}U$, (dots) $^{18}O + ^{208}Pb$). Curves represent estimates based on the liquid drop model: (dotted) $^{22}Ne + ^{232}Th$, (dashed-and-dotted) $^{22}Ne + ^{238}U$, (short dashes) $^{16}O + ^{232}Th$, (long dashes) $^{16}O + ^{238}U$, (heavy solid) $^{18}O + ^{208}Pb$. Thin lines are shown for clarity.

ions with strongly deformed actinide target nuclei. This yield was associated with the influence of closed shells $Z = 28$ and $N = 50$. The total kinetic energy of such fragments was higher than the predictions of the liquid-drop model. It was shown that the properties of mass–energy distributions exhibited features characteristic of modal fission for all of the studied reactions.

It was shown that in reactions of light ions with deformed actinide nuclei, the increase in the width of the mass distribution in the sub-barrier region results from the presence of the asymmetric modes in the compound nucleus fission. In addition, analysis of the experimentally measured cross sections of the formation of evaporative residues showed no deviations from the predictions of the statistical model. Since the increase in mass distribution variance and the high angular anisotropy of fragments can be readily explained using the concept of multimodal nuclear fission, there is no reason to expect the presence of orientation-dependent quasi-fission in the sub-barrier region for such systems.

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