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THE BINARY AND TERNARY DECAY OF HOT HEAVY NUCLEI PRODUCED IN THE REACTION ¹⁴N (34 AMeV) + ¹⁹⁷Au

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1. INTRODUCTION

Nuclear fission is the dominating decay mode for a large interval of excitation energies. This disintegration of heated heavy nuclei into two fission fragments (FF) of nearly equal mass mainly competes with the emission of neutrons and — at temperatures higher than 3 MeV [1] — light charged particles (LCP). Recently, a combined dynamical-statistical description of this complex interplay has been developed [2]. It is well established now that fission represents an overdamped collective motion over a saddle in the hyperplane of the potential energy to a considerably large deformed scission configuration and proceeds on a time scale of several units of 10^{-20} s [3].

The total kinetic energy release (TKE) of the fragments is defined by the Coulomb repulsion between the preformed FF's at the scission point. An empirical parametrization of the mean TKE was already given in 1966 [4] noting that the behaviour is explicitly governed by the Coulomb term $Z^2 / A^{1/3}$, where Z and A denote the charge and the mass numbers of the fissioning nucleus, respectively.

The emission of light particles from a heated nucleus, as treated by the statistical model, is usually considered to be an evaporation process. The probability P_{ev} is then given by the level density ρ , which for a Fermi gas takes the asymptotical form of a Boltzmann evaporation factor ρ (E*) ~ exp ($2\sqrt{a~E^*}$), where E* is the excitation energy, and the level density parameter - a - is proportional to A. In the case of LCP one has to take account of the Coulomb barrier (B_C) getting P_{ev} ~ exp ($2\sqrt{a~(E^*-B_C)}$). The characteristic time for particle evaporation can be evaluated by τ_{ev} ~ 1 / P_{ev} keeping in mind the statistical nature of the decay. The inclusive spectra of the particles are well described by Maxwellian distributions characterized by the temperature of the emitting nucleus. For *charged* particles, the spectra have a lower limit at B_C. Of course, the nucleus is no heat bath, but cools down during particle emission, what is essential in describing long evaporation cascades. The combined dynamical-statistical model of fission mentioned above is an attempt to take this feed-back into account in the fission-evaporation competition.

Investigations of heavy-ion induced reactions at intermediate energies — in the so-called Fermi-energy domain — which became possible in the 1980's, showed that besides

LCP's also complex fragments of intermediate mass (IMF's) are emitted. Somewhat arbitrarily the IMF's were defined as being fragments of mass $4 < M_{IMF} < 20 \div 30$ (or $2 < Z_{IMF} < 10 \div 15$) but, in any case, of mass between that of the evaporated LCP's and the FF's. They can originate from very different processes (cf. ref. [5]). For the present, we want to consider only such IMF's, which were emitted from an *equilibrated* (compound-like) source. The formation of an excited compound nucleus as a result of an incomplete fusion reaction, characterized by only partial linear momentum transfer (LMT < 1), has been observed in many experiments (e.g. refs. [6, 7]).

From pure statistical considerations, Moretto et al. [5] already supposed that "fission and evaporation are the two particularly (but accidentally) obvious extremes of a single statistical decay process, the connection being provided in a very natural way by the mass asymmetry coordinate". Since the transition-state model of fission delivers $P_f - \exp\left(2\sqrt{a^*(E^*-B_f)}\right)$ for the fission probability, i.e. an expression of the same form as for evaporation, the fission yield should – at sufficiently high E^* – only be governed by the energetically allowed phase space flux over the "ridge line" [8] – the line connecting the conditional saddle points (B_f) for all possible mass splits.

The statistical approach treating the disintegration of the compound nucleus as being controlled by the phase space only neglects, of course, any fission dynamics. The delay of the transition from saddle to scission [3], on the other hand, demonstrates the presence of dynamical hindrances mainly caused by the influence of the nuclear viscosity. It is, therefore, of interest to investigate how they affect other observables like, e.g., the TKE-M distribution.

In this work, we analysed the TKE-M distributions of the fragments generated in binary fission. The reaction 14 N (34 AMeV) + 197 Au [1] was used to produce equilibrated composite systems with excitation energies ≤ 400 MeV. This energy range is still dominated by the binary decay of the hot heavy nucleus [9, 10], which is accompanied by the emission of neutrons and LCP's during the de-excitation cascade. Nevertheless, the excitation energy is already sufficient for the decay into three massive fragments. In particular, a small amount of events is revealing the occurrence of an IMF measured in coincidence with two FF's [12, 13]. The origin of these IMF's is a further interesting question. Especially, the time evolution of the disintegration process is essential. If the

IMF was emitted well before fission starts, both the excitation energy and the fissility of the heavy remnant were much more reduced than in the case of a prior-to-fission emitted light particle. A time-scale analysis of three-fragment decays of a composite system produced in the reaction ²²Ne (60 AMeV) + ¹⁹⁷Au was performed in ref. [14] by considering angular and velocity correlations. The best agreement between the data and the results of trajectory calculations was obtained there, if a rather fast sequential process has been assumed. The mean time interval between the two fragment separations turned out to be 10⁻²¹ s.

Another distinct low-energy IMF-component was found in ref. [15]. Because of the focusing of that yield into angles near 90° with respect to the fission axis, the effect was interpreted as emission out of the neck formed during fission.

In the reaction 14 N (34 AMeV) + 197 Au, we recorded also three-fragment events. We performed a correlation analysis, which is especially sensitive to the time interval between the IMF emission and the final scission of the system. Using the limited statistics of the present experiment, however, only a qualitative discussion is possible. A more detailed analysis of three-fragment correlations is planned to be performed on the basis of a high-statistics data body recently recorded for the reactions 14 N (53 AMeV) + 197 Au and 14 N (53 AMeV) + 232 Th.

2. THE EXPERIMENTAL METHOD

The measurement has been carried out at the heavy-ion beam of the U-400M cyclotron of the FLNR at the JINR Dubna using the 4π -fragment spectrometer FOBOS [16]. This multi-detector array consists of 30 combined detector modules mounted on the facets of a truncated icosahedron and realizing a so-called logarithmic detector device.

Two detector shells, namely, i) position-sensitive avalanche counters and ii) axial-field (Bragg) ionization chambers measure the coordinates (\mathfrak{D}, φ) , the time-of-flight (TOF), the residual energy (E) and the Bragg-peak height (BP ~ Z) of the fragments, whereas the third shell iii) consisting of CsI(Tl) scintillators is especially suited for the LCP identification by use of the pulse-shape analysis method [17].

From the measured quantities, the individual fragment masses (M_F) and the momentum vectors (P_F) can be derived applying the TOF-E method "event-by-event" without any kinematical assumption [18]. For two-fragment decays, the sum of the parallel momenta ($P_{F1} + P_{F2}$)|| was checked to select events of large LMT \approx 0.8. Furthermore, in order to ensure the completeness of the massive fragment detection, the sum of the fragment masses was analysed. The LMT has been used as a rough measure of E* of the composite system. A sufficiently large value of the total fragment mass ($M_{F1} + M_{F2}$) together with a limited transverse momentum ($P_{F1} + P_{F2}$) $_{\perp}$ < 500 MeV/c were used as criteria for the selection of coplanar binary decays. The TKE was calculated from the both independently measured masses and fragment velocities. This method avoids any influence of pre-scission processes (fluctuations in incomplete fusion and in the evaporation cascade) on the result.

We must emphasize here that – in the very asymmetric reaction induced by the light ^{14}N projectile – fragments of $M_F \ge 14$ should only originate from the *decay of a compound-like system* and deep-inelastic components are strongly suppressed. In reactions induced by heavier projectiles (like ^{40}Ar , ^{27}Al ; see refs. [9, 19]), this is generally not the case and the picture becomes more complicated. An additional condition for ruling out any possible fast processes was the selection of only such events for the further analysis, where the lighter of the both fragments was emitted "backwards" in the c.m. frame.

At energies of $E^* \le 400$ MeV of the hot system produced in the given reaction, the amount of three-body decays (IMF-accompanied fission) is less than 1 % [13] and the bulk of the data is due to binary disintegrations. The three-fragment events were checked by the same criteria as in the binary case, but the sums were taken over three fragments and the entire LMT-range was accepted.

A special method has been applied to study proximity effects in IMF-accompanied fission. The c.m. velocity ($\nu_{F1 F2}$) of the two heavy fragments (F1, F2) was determined from both their masses and momentum vectors by eq. 1 and the velocity (ν^{lab}) of the third fragment (IMF) was then transformed by eq. 2 into this frame (ν^{rel}):

$$v_{F1 F2} = (P_{F1} + P_{F2}) / (M_{F1} + M_{F2})$$
 (1)

$$v^{\text{rel}}_{\text{IMF}} = v^{\text{lab}}_{\text{IMF}} - v_{\text{F1 F2}} \tag{2}$$

The angle between the direction of the emitted IMF and the fission axis with respect to (F1, F2) was determined in the same frame.

3. TWO - FRAGMENT DECAY

3.1 Experimental results

Binary events restricted by the above formulated conditions are shown in the TKE versus M contour plot of fig. 1. To demonstrate the large width of this distribution in mass and energy and to illustrate the resolution obtained by the application of the TOF-E method, we chose a logarithmic intensity scale with a factor of 2 between subsequent contour lines.

The main yield in fig. 1 is due to normal symmetric multi-chance fission of the hot equilibrated system, but very asymmetric mass splits extend to fragment pairs usually classified by their masses as IMF's and heavy residues, respectively. The mean value $\langle M_F \rangle = 176$ a.m.u. corresponds to an average mass-loss (with respect to complete fusion) of 35 a.m.u. due to pre-compound particle emission (incomplete fusion) as well as pre- and post-scission evaporation. The branch of the heavy fragment is slightly broader than that of the light one because of the larger corrections for energy losses in the detector window materials and, therefore, slightly larger uncertainties in the mass determination are observed.

The large TOF-path of the FOBOS array (50 cm) and the timing properties of the position-sensitive avalanche counters allow an accurate measurement of the fragment velocities (ν_F). The deduced relative velocities between binary fragment pairs (ν_{rel}) are shown in dependence on ν_F in the contour plot of fig. 2. The mean value at symmetric fission of $\nu_{rel} > \nu_{rel} > \nu_{rel} > \nu_{rel} = 2.4$ cm/ns is in accordance with the systematics of ref. [4].

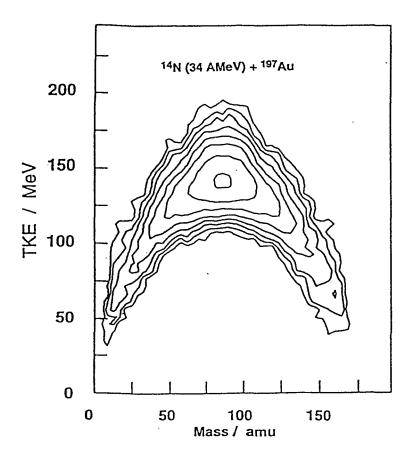


Fig. 1 TKE-M distribution of binary fragment pairs of the hot compound system formed after incomplete fusion (LMT \approx 0.8) in the reaction 14 N (34 AMeV) + 197 Au [20].

By scaling of the TKE formula [4] with the asymmetry factor $4M_1 M_2 / (M_1+M_2)^2$, accordance of the experimental data $< v_{rel} >$ with the derived values is observed for asymmetric mass splits down to about 1:3. At larger mass asymmetry of the decay, the $< v_{rel} >$ considerably deviate from a parabola, as shown in fig. 2.

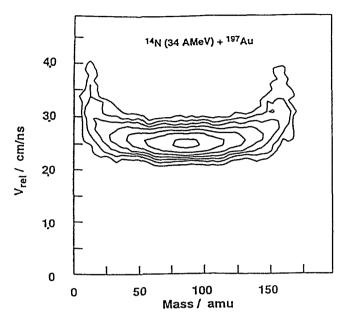


Fig. 2 v_{rel}-M distribution for the same events as in fig. 1 [ref. 20].

A similar deviation of measured $< v_{rel} >$ from a Coulomb calculation was earlier observed for asymmetric binary decays in the reaction 139 La $(18 \text{ AMeV}) + ^{12}$ C (cf. fig. 23 in ref. [5]). There, the $< v_{rel} >$ were found to be increasingly larger than the calculated values with decreasing charge number of the fragments starting at Z < 20. Our observations agree with this result qualitively.

3.2 Analysis of the TKE - M distribution

Using the data presented in figs. 1 and 2, we analyzed the TKE spectra for mass bins of $\Delta M_F = 5$ a.m.u. These spectra have a symmetric shape except for the smallest fragment masses at $M_F < 25$ a.m.u. The mean values <TKE> are plotted versus the mean values of the mass bins in fig. 3.

The $\langle TKE \rangle$ and the standard deviations σ (TKE) were determined by Gaussian fits over ranges in these spectra, where the yield exceeds 10 % of the maximum value. For comparison, we show also the calculated TKE values [4] and the Coulomb barrier B_C [21].

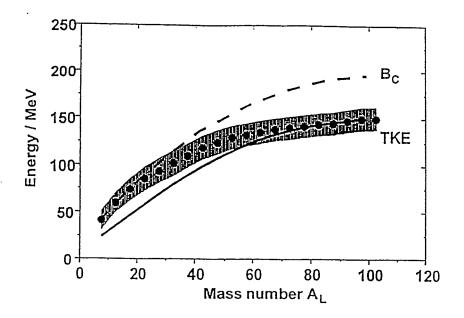


Fig. 3 Measured <TKE> versus the fragment mass (full circles). The hatched area corresponds to $\pm \sigma$ (TKE). The full line is calculated using the TKE formula [4] scaled for asymmetric mass splits. The dotted line represents a B_C-calculation.

Starting from symmetric fission, one observes that the <TKE> – the "most probable" TKE values for the mass bins considered – at first follow the line calculated by use of the TKE formula and then smoothly approach the B_C -line. Below $M_F \approx 50$ a.m.u., the deviation from the calculated TKE values exceeds $\sigma(TKE)$ and below $M_F \approx 25 \div 30$ a.m.u. the <TKE> are well reproduced by the Coulomb barrier B_C .

Assuming for fission of hot nuclei that $M_1/M_2 = Z_1/Z_2$ (where the compound nucleus is given by $M = M_1 + M_2$ and $Z = Z_1 + Z_2$), the scaling factor for the calculation of the TKE values at asymmetric mass splits can be taken as $4M_1 M_2/(M_1+M_2)^2$ or $4Z_1 Z_2/(Z_1+Z_2)^2$. It is obvious that, in this manner, one takes only account of a redistribution of the charge and mass numbers of the fissioning nucleus between the fragments. In the framework of the two-spheres approximation [4], the Coulomb repulsion at scission is responsible for the TKE release and changes with the effective distance D_{sc} between the fragments. Formally, one gets $D_{sc} \sim A_1^{1/3} + A_2^{1/3} \le (A/2)^{1/3} \sim D_{sc}^{sym}$. This approximation does not hold for more asymmetric mass splits.

Consequently, the average scission shapes should become more compact leading to an enhanced Coulomb repulsion and, therefore, to the larger <TKE> values observed in this work (fig. 3).

This behaviour of the <TKE> reflects the approach of the conditional scission points to the ridge line of conditional saddle points with increasing asymmetry of the binary decay. Furthermore, as the descent from the saddle to the scission point is responsible for a large contribution to the fission transient time [3], this should be a hint that more asymmetric disintegrations proceed faster than symmetric fission because of their minor damping.

If we understand the difference between the barrier B_C and the measured <TKE> as to be the mean amount of dissipated energy (E_{Diss}) on the fission path to scission, the vanishing damping at sufficiently large mass asymmetry becomes evident. With the expression $A_1 A_2 / A^2$ chosen for the mass asymmetry, the dependence of the dissipation on this parameter turns out to be linear in a fairly broad region (fig. 4). For the most asymmetric mass splits, E_{Diss} becomes formally even negative reflecting the amount of kinetic energy, which the light cluster gets from the hot emitting nucleus in an evaporation process.

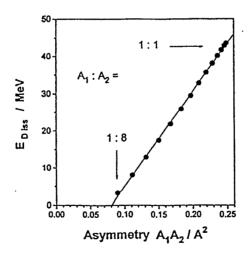


Fig. 4 Mean dissipated energy $E_{Diss} = B_C - \langle TKE \rangle$ in dependence on the mass asymmetry expressed by $A_1 A_2 / A^2$.

4. THREE - FRAGMENT DECAY

4.1 Experimental results

From the 1200 three-fragment events recorded in this experiment, we estimated an integral ternary to binary decay ratio of $4\cdot10^{-3}$ for the reaction ^{14}N (34 AMeV) + ^{197}Au . The included correction for the geometrical acceptances leading to different registration efficiencies for binary and ternary events are based on a Monte-Carlo simulation.

The spectra of the relative velocities between the IMF (A = $10 \div 20$) and, either the heavy partner in a binary decay (v_{rel}^{bin}), or the center-of-mass of the two heavy fragments in a ternary decay (v^{rel}_{IMF}) are shown in fig. 5.

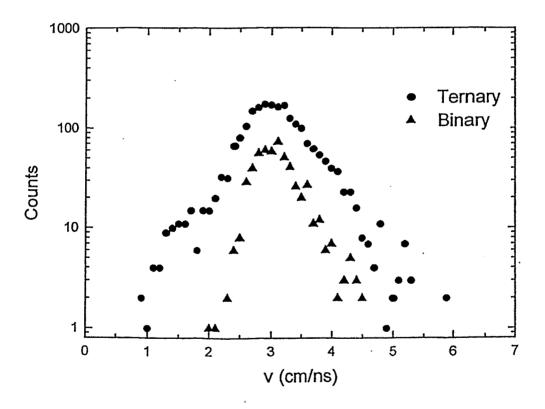


Fig. 5 Relative velocities between IMF's of $10 \div 20$ a.m.u. and their heavy partners in asymmetric binary decays (v_{rel}^{bin}) and between IMF's and the center-of-mass of the two fission fragments in ternary decays (v^{rel}_{IMF}).

The peaks in the two spectra correspond relative to their position. Furthermore, a second component at lower velocity is evident in the v^{rel}_{IMF} - distribution. In ref. [15], such a low-energy component was interpreted as an IMF-emission out of the neck region of the fissioning nucleus, where the Coulomb repulsion is reduced. In this case, some Coulomb focusing should be observed and, therefore, the ratio of the low-velocity to the high-velocity IMF-yields versus the emission angle with respect to the fission axis was plotted in fig. 6. Here, effects due to geometrical acceptances are excluded. A certain enhancement near 90° is really observed, but some events are also observed at other angles.

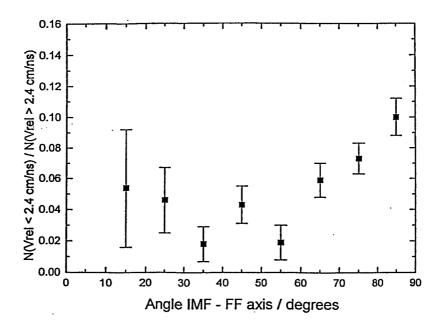


Fig. 6 Angular distribution of low-velocity IMF's with respect to the fission axis. [24]

The influence of a third fragment (IMF) on the relative velocity between the two fission fragments is demonstrated in fig. 7. In those events, where the IMF's have a high velocity, the FF's have a mean relative velocity of 2.4 cm / ns as expected for a usual fission process [4]. The emission of an IMF with low velocity, on the other hand, leads to a remarkable enhancement of the relative velocity between the remaining two heavy fragments.

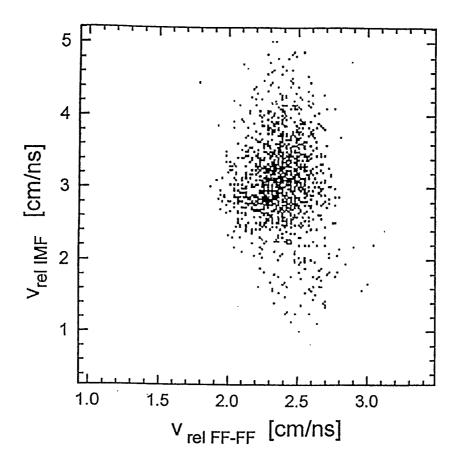


Fig. 7 Scatter plot of the IMF velocity relative to the fission fragment c.m. frame versus the relative velocity between both fission fragments in ternary events.

The yields of the both ternary components per binary fission are shown in fig. 8 in dependence on the LMT determined from the sum of the momenta of the three fragments. The yield of the high $v^{\rm rel}$ $_{\rm IMF}$ -component strongly increases with increasing LMT, whereas the low-velocity component remains almost constant.

The Z-distributions of IMF's emitted with high and low relative velocities, respectively, are compared in fig. 9. The high-velocity component decreases much stronger with increasing Z than the low-velocity one. The second component shows also an odd-even effect up to Z = 10.

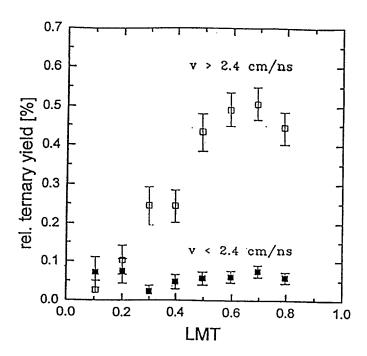


Fig. 8 Yields of the two components of ternary decays per binary fission in dependence on the transferred linear momentum. [24]

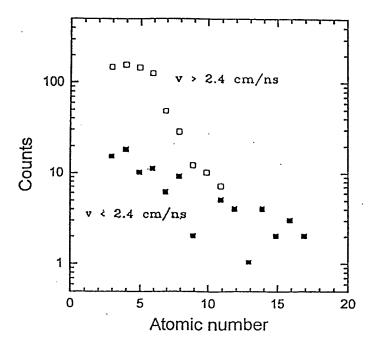


Fig. 9 Z-distributions of IMF's observed in ternary events at low and high relative velocities. [24]

4.2 Discussion of the ternary decays

The correspondence of the peak positions of the high- v^{rel} $_{IMF}$ component in ternary decays with that of the relative velocities between IMF's of comparable mass and the heavy remnants in binary events (v_{rel} bin ; fig.5) supports the suggestion that both components have the same origin. The only difference is that the heavy remnant, which remains after the IMF was emitted, might subsequently undergo fission or not. This means that in the three-fragment decay the fission process occurs later, and does not influence the IMF velocity.

As the IMF needs about $3 \cdot 10^{-21}$ s for being accelerated to ≈ 80 % of its asymptotic velocity by the influence of the Coulomb force, it can be concluded that the time interval between the IMF emission and the subsequent fission amounts to at least several units of 10^{-21} s. Consequently, the removing IMF left only a lighter and less excited nucleus, but beyond it, did not influence the subsequent fission process. This suggestion is also in agreement with the observed relative velocity between the two FF's (fig. 7), which turns out to be the same as in a binary decay. Such IMF-accompanied ternary events are of clear sequential nature — i.e. the IMF is "prior-to-fission"- emitted.

On the other hand, we interpret such ternary decays, which are distinguished by an IMF with lower velocity, as fission combined with a neck-emission. The neck region of the fissioning nucleus should be regarded as possible source of these IMF's not only because of the Coulomb focusing effect (cf. ref. [15]), but - beyond this - also due to the present observation of an *increased relative velocity of the FF's* (fig. 7). A third fragment, when created "between" the two separating FF's, influences the Coulomb repulsion. Under the assumption that roughly the total (potential) Coulomb energy of the three nearby-formed fragments in a ternary decay is transformed into kinetic energy, a decrease in the kinetic energy of the IMF should lead to an increase of the kinetic energy of the FF's. Quantitative conclusions, of course, will only be possible by the comparison with trajectory calculations planned for the next period.

Such calculations should also clarify the origin of the observed low-velocity, but *non-focused*, IMF's. Possible scenarii could be, e.g., : i) no emission from the neck, but out of the deforming nucleus during fission, when the Coulomb barrier is lowered, or ii) a slightly delayed second neck-rupture between the nascent fragments.

There is a striking difference between the excitation functions (fig. 8) of the two IMF components discussed (assuming the LMT as a measure of E*). Such a behaviour has already been observed in the analysis of IMF-accompanied fission in the reaction ⁷Li (43 AMeV) + ²³²Th [22]. This fact can be interpreted as a consequence of the dynamics of the fission process. If the emission times are different, it should be obvious that the "early-emitted" high-velocity component is more affected by the primary excitation energy (E*) of the composite system than the neck component. The systematics of the excitation energies remaining in the FF's [3] shows a very weak dependence on E*. Consequently, the excitation energy of the fissioning system near scission should also depend only weakly on E*. Thus, a nearly constant yield of the neck component with LMT results in fig. 8.

The odd-even effect obvious in the Z-distribution of the low-velocity component (fig. 9) is a further hint that the excitation energy near scission is rather small. The Z-distribution of the prior-to-fission emitted IMF's does not show any odd-even effect and drops steeply at $Z_{IMF} > 6$. These IMF's emitted at high excitation energies – i.e. in an early stage of the de-excitation process – reduce progressively the fission probability of the heavy remnant with increasing Z_{IMF} and the less fissile and less excited remnant has increasingly the chance to survive as a heavy residue. This means that early-emitted IMF's of large Z_{IMF} "favour" the binary decay.

In this context, there is an interesting connection with the observations discussed above concerning very asymmetric binary decays. Namely, extrapolating the steep slope of the Z-distribution of the prior-to-fission emitted IMF's (fig. 9) to zero and assuming $A_{IMF}=2~Z_{IMF}$, a mass number of $A_{pre}^{max}\approx26~a.m.u.$ is obtained. This is roughly the mass region, where the <TKE> of very asymmetric binary decays approaches to the Coulomb barrier B_C (fig. 3). This behaviour can be interpreted as the gradual disappearance of the dissipation during the disintegration process. The extrapolation of the dependence in fig. 4 gives A_{ev} ($E_{Diss}=0$) $\approx15\div16~a.m.u.$ for the system considered. It can be assumed approximately that light "clusters" of mass up to an A_{ev} coresponding to $Z_{ev}\approx7\div8$ can be evaporated by the hot compound-like nucleus during the de-excitation cascade. Indeed, the steep slope of the Z-distribution of the

prior-to-fission emitted IMF's (fig. 9) begins at $Z_{IMF} = 7$ and the yield at $Z_{IMF} \le 6$ is rather constant.

Keeping in mind that the prior-to-fission IMF component is not affected by the later occurring fission process and, therefore, supposed to be emitted "earlier" (i.e. at high E^*), we can assume that $E^* >> B_C$ and the probability $P_{ev} \sim \exp\left(2\sqrt{a(E^*-B_C)}\right)$ is reduced (by neglecting phase space constants) formally to $P_{ev} \sim \exp\left(2\sqrt{a(E^*)}\right) = f(E^*)$.

Starting from some critical A^{max} , dynamical considerations come into play and the IMF emission loses its statistical feature and follows now a dynamical time scale. This means that the nature of the decay process changes from *evaporation-like* to *fission-like* [5]. The more asymmetric the mass split is, the lower is the dissipation (fig. 4) and - most probably - the faster is the disintegration. The drop of the yield of the prior-to-fission IMF component at some Z_{ev}^{max} is in agreement with such a scenario. Of course, at higher incident energies than in the reaction considered, the yields of ternary IMF's with higher Z_{IMF} should increase and the decay mechanism should develop from (sequential) IMF-accompanied fission to the limit of ternary fission [23]. This process should be governed by the dynamics of the collective motion of the nuclear matter involved.

From energy considerations, namely, that the fission barrier increases, but the Q-value of reaction decreases with increasing mass asymmetry of the binary decay, the disintegration into very asymmetric fragments carrying away a TKE > Q needs, on principle, a larger amount of E* to occur than the symmetric fission of the same system. This means that the effect of the intrinsic single-particle motion on the collective degrees of freedom, which is responsible for a fission-like process, should be temperature-dependent. More asymmetric modes are only generated at sufficiently large E*, or fission at asymmetric mass splits should proceed faster, i.e. at a time scale, when the system has not yet been cooled down considerably by particle evaporation. Up to now, there is no consistent description of the complex interplay of light-particle as well as IMF evaporation and fission into the broad range of mass splits observed experimentally. The method of ref. [2], which combines statistical as well as dynamical aspects of this process, should presently be the most adequate one, but it has to be extended by including of more degrees of freedom representing a very complicated task.

Moreover, the broad Z-distribution observed for IMF's emitted from the neck cannot be explained by simple assumptions about excitation energies, emission barriers, etc. Up to

now, there is no consistent theory describing the neck emission of IMF's in the given energy range. Probably, it is also governed by the complex dynamics of the fission process including the formation of the scission configuration and the rupture of the neck.

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