Isotopic composition of fragments in nuclear multifragmentation

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In this Rapid Communication we present results from the analysis of the isotopic yields of fragments emitted in two selected reactions: the decay of the quasiprojectile in Au+Au peripheral collisions at 35 MeV/nucleon, and the disassembly of the unique source formed in Xe+Cu central reactions at 30 MeV/nucleon. We find that the relative yields of neutron-rich isotopes increase with the excitation energy of the emitting sources. In the framework of a statistical multifragmentation model which reproduces fairly well the experimental observables, such behavior can be explained with the increase of the $N/Z$ ratio of the hot primary fragments. This corresponds to the statistical evolution of the fragmentation mechanism as a function of the excitation energy, from the decay into few small fragments with a heavy residue to complete multifragmentation.

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Nuclear fragmentation and its connection to the behavior of nuclear matter at high excitation energy is the subject of intensive theoretical and experimental investigations [1]. Some general properties of this process are already established: at relatively small excitation energies ($E^* \approx 2\text{–}3 \text{A}\text{MeV}$) there is a formation and decay of a long-lived compoundlike nucleus system. This process can be described by evaporation/fissionlike models. At higher excitation energies (close to the binding energy) there is a complete fast disintegration of the system into fragments. This process can take place in a finite breakup volume. In this case statistical models based on the hypothesis of a nuclear phase transition (simultaneous decay) happen to be very successful [2,3].

In this Rapid Communication we present recent data on the isotope production in heavy-ion collisions at intermediate energies [4,5] with the aim of studying the isotopic content of fragments for different sources and excitation energies during the transition from the low energy decay to the multifragmentation. We will show that the behavior of the experimental isotopic yields, as a function of source size, isospin, and excitation energy, can be connected to the corresponding evolution of the $N/Z$ ratio of the hot primary fragments in the breakup volume, which is an important ingredient in the disintegration process.

In fact, there are both experimental and theoretical studies which show that the knowledge of the chemical composition of hot fragments helps in establishing the freeze-out conditions.

(1) Information about the density of the freeze-out volume can be obtained from the analysis of the velocity correlation functions and kinetic energies of the emitted fragments. Among the different methods [6,7] to extract the temperature of the system, the most popular is based on the statistical properties of the double isotope ratios. With this technique a nuclear caloric curve, as an experimental evidence of a nuclear liquid-gas phase transition [8], was obtained. In any case, a correction for secondary decay of hot fragments produced in the freeze-out volume [9] is needed, and depending on their $N/Z$ ratio, the secondary decay can proceed differently.

(2) An original method to investigate thermodynamical conditions at the freeze-out via the energy and particle balance was proposed in [10]. To apply this method one needs to know about the chemical composition of primary fragments.

(3) In order to extract excitation energies of hot primary fragments, a sophisticated analysis of fragment correlations was suggested [11]. Depending on the $N/Z$ ratio of the primary fragments this method leads to different results.

(4) On the theory side, the chemical composition of hot fragments is directly related to the symmetry energy term of the nuclear equation of state at subnuclear densities. Furthermore, it plays a role in establishing the difference between the dynamical and statistical mechanisms of fragmentation [12].

We investigated the Au+Au at 35 MeV/nucleon and Xe+Cu at 30 MeV/nucleon reactions. The experiments were performed at the National Superconducting K1200 Cyclotron Laboratory of the Michigan State University. The angular range $3^\circ < \theta_{\text{lab}} < 23^\circ$ was covered by the MULTICS array [13]. The identification thresholds in the MULTICS array were about 1.5 MeV/nucleon for charge identification and about 10 MeV/nucleon for mass identification. The MULTICS array consisted of 48 identical telescopes, composed of an ionization chamber (IC), a silicon detector (Si), and a CsI crystal. Typical energy resolutions were 2%, 1%, and 5% for IC, Si, and CsI, respectively. Light charged particles and fragments with charge up to $Z = 20$ were detected at $23^\circ < \theta_{\text{lab}} < 160^\circ$ by the phoswich detectors of the MSU Miniball hodoscope [15]. The charge identification thresholds were about 2, 3, 4 MeV/nucleon in the Miniball for $Z$
=3, 10, 18, respectively. The geometric acceptance of the combined array was better than 87% of \( 4\pi \).

The reduced impact parameter \( \hat{b} \) is determined by the following expression [14]:

\[
\hat{b} = b / b_{max} = \left( \int_{Nc}^{+\infty} P(N')dN' \right)^{1/2},
\]

where \( P(Nc) \) is the charged particle probability distribution and \( \pi \cdot b_{max}^2 \) is the measured reaction cross section for \( Nc \) \( \geq 3 \).

The fragments coming from the decay of the quasiprojectile in peripheral \( \text{Au} + \text{Au} \) 35 MeV/nucleon reaction and those coming from the disassembly of the unique source formed in \( \text{Xe} + \text{Cu} \) 30 MeV/nucleon central collisions have been identified through a careful data selection, taking into account possible distortions on energy and angular distributions due to the experimental efficiency [4,5]. In particular it was verified that all the detected decay products are emitted nearly isotropically from the same source and that their energy distributions have Maxwellian shapes. In both measurements, angular and energy distributions are compatible with statistical emission, providing an experimental indication that thermalization of the emitting sources has been reached [4,5].

The reconstruction of the excitation energies of the sources was carried out analyzing the kinematical characteristics of the produced fragments (calorimetric evaluation [10,16]). For the reaction \( \text{Au} + \text{Au} \) we selected impact parameters \( 0.95 > \hat{b} > 0.5 \), which give for quasiprojectile \( \text{Au} \)-like sources excitation energies from 3 to 6 MeV/nucleon, while for the \( \text{Xe} + \text{Cu} \) reaction we choose \( \hat{b} < 0.2 \), which provide fusion sources with excitation energy around 5.5 MeV/nucleon. In such a way we select two sets of emitting sources with similar \( A \), but different \( Z \). In Refs. [4,5] these data were used to obtain a caloric curve with the double isotope ratio method.

Here we wish to present further information about the composition of hot fragments, which require the study of new observables, such as the isotopic yields for fixed elements and their evolution with the excitation energy and other parameters of the emitting source. The analysis of the relative isotope production can provide more reliable information about the statistical picture of the process than the analysis of the isobars. In fact, the neighboring isobars (with \( \Delta Z = 1 \)) can be produced at different Coulomb barriers (up to 10 MeV for the \( \text{Au} \) source). The uncertainty in accounting for the real Coulomb energy of the isobars at freeze-out may essentially exceed the difference in their binding energy \((\sim 1 \text{ MeV})\) rendering it impossible to unambiguously determine the thermodynamical parameters.

In order to avoid a possible problem of preequilibrium in the emission of light charged particles we mainly focused on the analysis of IMFs. In Fig. 1 we show the relative isotopic yields versus excitation energy obtained from the experimental data for \( \text{Au} \) sources (each isotopic yield is normalized to the total yield for fixed \( Z \) value). One can see a trend: The relative isotopic yields of neutron rich fragments increase with the excitation energy. In Fig. 2 we present the ratios of yields of measured isotopes with the largest and smallest number of neutrons at fixed \( Z \) values, versus the excitation energy. For all analyzed IMFs, the ratio increases considerably in the energy range \( E^* = 3–6 \text{ MeV/nucleon} \). As was shown in Ref. [4] (see evolution of charge distributions in their Fig. 8) the lowest energy of this range corresponds to the onset of multifragmentation with mean IMF multiplicity around one plus a heavy residue, while at the highest excitation energy the decay into many IMFs dominates. In this respect our data cover the energy range where a phase tran-
sion could take place [10]. At the excitation energy of about 5.5 MeV/nucleon we found that the abundances of neutron-rich isotopes are larger for the peripheral quasiprojectile Au than for the central Xe + Cu unique source. This trend has a natural explanation since the $N/Z$ ratio of the Au source is larger than the Xe + Cu one. The relative yields at this excitation energy are shown in Fig. 3. The observed trends would be consistent if the $N/Z$ ratio of the primary fragments increases with the excitation energy of the sources.

Several experimental and theoretical results have shown that in the studied regime the disassembly of the emitting sources can be described in terms of statistical models (see, e.g., [10]). To better understand the $N/Z$ ratio behavior with the excitation energy, we decided to refer to the statistical multifragmentation model (SMM) [2], which reproduces the observed charge yields, the He-Li isotope temperatures [4,5], as well as other observables. In the following we use the set of SMM parameters which gives the best description of multifragmentation of the sources produced in peripheral collisions [4,10]. The freeze-out density was taken $1/3 \cdot \rho_0$ ($\rho_0$ is the normal nuclear density). The hot fragments are described in the liquid-drop approximation with the Coulomb interaction [2]. The crucial condition for the present isotope analysis is the requirement of a full description of the yields for each considered charge distribution. Under this condition the same parametrization for the central Xe + Cu reaction was used. A possible slight decrease of the source size, as a result of preequilibrium emission, does not affect the conclusions because it hardly changes the $N/Z$ ratio of the source [2]; likewise a small dynamical expansion effect has minor importance. We checked that the calculated isotopic trends (see below) remain stable with respect to reasonable variations of the SMM parameters in the ranges where the charge yields and other observables are reproduced.

Comparison of the SMM predictions with the data is shown in Figs. 1–3. The qualitative agreement is evident (even quantitative for some important isotopes) and the general trends, especially the increase with excitation energy of the neutron-rich isotopes with respect to the neutron-deficit ones, are correctly reproduced. Some discrepancies in the results can be ascribed to approximations in the calculations. In the SMM, Fermi breakup is used to describe the secondary decay of fragments with $A \approx 16$ [17]. The model takes into account all ground and nucleon-stable excited states of light fragments and calculates the probabilities of population of these levels microcanonically (according to the available phase space). It does not include matrix elements of the transitions between these states that can be important at small excitation energies. Also the model does not take into account for possible shifts of the nuclear states caused by Coulomb interaction of the excited fragments with the surrounding nuclear matter. These shifts should be calculated in consistent quantum theories. However, in our cases we expect a rather high excitation energy of hot primary IMFs, considerably larger than thresholds of the main breakup channels ($\approx 2$ MeV/nucleon), and the above mentioned problems do not affect the calculated trends.

According to the SMM predictions [2], at the beginning of multifragmentation the number of primary nucleons in the freeze-out is very small and nearly all available protons and neutrons are bound in hot primary fragments. Their $N/Z$ ratio depends on the fragment size. If there are light and heavy fragments in the freeze-out, the light fragments have typically smaller $N/Z$ ratio than the heavier ones: These channels are more energetically favorable because of an interplay between the symmetry and the Coulomb terms [17]. In Fig. 4 we show how the mean $N/Z$ ratios for the light ($Z=8–10$) and heavy ($Z=68–70$) primary fragments evolve with the excitation energy. It is interesting to note that, if a residue-like hot fragment is present in the freeze-out, its $N/Z$ ratio can be even larger than the corresponding source ratio because the other light fragments have a considerably lower ratio. At excitations higher than the multifragmentation threshold ($E_{\text{th}}=3–4$ MeV/nucleon), heavy fragments decrease in size, and therefore, more neutrons are combined
into hot IMFs. Finally, at very high excitations ($E^* \sim 8$ MeV/nucleon) the $N/Z$ ratio of the hot IMFs decreases, because the fragments larger than IMFs are no longer present, and the number of primary free neutrons increases fastly. As a consequence of this evolution, we observe an increase of the $N/Z$ ratio of the cold fragments in the energy range $E^* = 3–6$ MeV/nucleon, and a sizable change in the relative yields of neutron-rich and neutron-deficit isotopes (see Figs. 1, 2). At higher excitation energies the calculated ratio will drop (see $Z=5$ in Fig. 4) similarly to the hot fragment one.

Nevertheless, there are alternative ways to produce IMFs. For instance, according to SMM at small excitation energy, light IMFs can be emitted from large fragments via the evaporational mechanism [17], which favors the production of nearly symmetric isotopes with large binding energies (close to $\beta$-stability line). As a consequence, their $N/Z$ ratios are usually smaller than the ratio of the source, and even though the probability of the evaporation of IMF is small, it contributes to the observed yields at $E^* \leq E^*_{thr}$. At excitation energies near the threshold, the multifragmentation sets in: It evolves from a fast breakup in two hot fragments, which looks like an evaporational channel, towards the breakup into three or more fragments with the increase of the source excitation energy [2]. In the multifragmentation regime, the secondary decay of hot primary fragments is the leading process, defining a relative abundance of particular isotopes. Switching off the multifragmentation and allowing only the evaporation process from the source, one cannot reproduce the charge yields, neither to explain the observed trend of the ratio at high excitation energies. Moreover, in the studied excitation energy range the relative contribution of the evaporated isotopes becomes negligible. Therefore, the evolution of the isotopic composition of hot IMF predicted by the SMM is fundamental to explain the observed effect.

In other experimental works, e.g., Ref. [18], it was similarly found that an increase of the $N/Z$ source ratio leads to increasing the relative yields of neutron-rich isotopes. A possible production of neutron-rich hot primary fragments is also reported in other models: Dynamical stochastic mean field calculations [12] for Au source at high excitations predict hot fragments with the same $N/Z$ ratio as the SMM. An analysis of the correlation functions performed in [11] reaches the conclusion that a larger neutron content of the hot primary fragments can explain their experimental data.

In summary, we presented new data on isotopic yields produced after decay of the Au and Xe+Cu sources in the excitation energy range 3–6 MeV/nucleon, around and slightly above the multifragmentation threshold. We found that the experimental relative yields of neutron-rich isotopes increase with excitation energy for the Au sources. This behavior is well reproduced by the SMM calculations and consistent with the statistical picture realized in the model. In this approach, the energy dependence of the isotopic composition of the produced fragments can be explained in terms of the evolution from a decay into few light fragments and a heavy residue, to the total multifragmentation breakup. This process can lead to an increase of neutron content of hot primary intermediate mass fragments.

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