PHYSICAL REVIEW C **66**, 021601(R) (2002)

Isotopic composition of fragments in multifragmentation of very large nuclear systems: Effects of the chemical equilibrium

P. M. Milazzo,[1] A. S. Botvina,[2] G. Vannini,[3] M. Bruno,[3] N. Colonna,[4] M. D'Agostino,[3] F. Gramegna,[5] I. Iori,[6] G. V. Margagliotti,[1] P. F. Mastinu,[5] A. Moroni,[6] and R. Rui[1]

[1]*Dipartimento di Fisica and INFN, I-34127 Trieste, Italy*

[2]*Gesellschaft fur Schwerionenforschung, Darmstadt, Germany and Institute for Nuclear Research, Russian Academy of Science, RU-117312 Moscow, Russia*

[3]*Dipartimento di Fisica and INFN, I-40126 Bologna, Italy*

[4]*INFN, I-70126 Bari, Italy*

[5]*INFN, 35020 Laboratori Nazionali di Legnaro, Italy*

[6]*Dipartimento di Fisica and INFN, I-20133 Milano, Italy*

(Received 12 February 2002; published 21 August 2002)

Studies on the isospin of fragments resulting from the disassembly of highly excited large thermal-like nuclear emitting sources, formed in the ^{197}Au+ ^{197}Au reaction at 35 MeV/nucleon beam energy, are presented. Two different decay systems (the quasiprojectile formed in midperipheral reactions and the unique source coming from the incomplete fusion of projectile and target in the most central collisions) were considered; these emitting sources have the same initial N/Z ratio and excitation energy ($E^* \simeq 5-6$ MeV/nucleon), but different size. Their charge yields and isotopic content of the fragments show different distributions. It is observed that the neutron content of intermediate mass fragments increases with the size of the source. These evidences are consistent with chemical equilibrium reached in the systems. This fact is confirmed by the analysis with the statistical multifragmentation model.

DOI: 10.1103/PhysRevC.66.021601 PACS number(s): 25.70.Pq, 24.60.−k

The study of heavy-ion collisions at intermediate energies ($10 \leqslant E \leqslant 100$ MeV/nucleon) is a useful tool to investigate the mechanisms of fragment production in highly excited nuclear systems. In this energy regime multifragmentation appears as one of the main deexcitation channel [1], which can be considered [2,3] as a manifestation of a liquid-gas type phase transition in finite nuclear systems. This gives an access to the nuclear equation of state. It has been shown that statistical models [4,5] happen to be very effective in the reproduction of main characteristics of the fragment production such as charge distributions and fragment correlations.

Nowadays, with the advent of radioactive beam facilities, the influence of the isospin degree of freedom is strongly addressed and experimental information on the isotopic content of emitted fragments represents a meaningful starting point in order to get a deep understanding of either the deexcitation mechanisms or the nuclear matter properties.

Theoretical calculations predict that, in the fragmentation of asymmetric nuclear matter, the isospin composition of the "liquid" phase [usually associated with intermediate mass fragments (IMF) and heavy residues] and the "gas" phase [light charged particles (LCP) and nucleons] depends on many factors. In particular, calculations by Müller and Serot [6] showed that, for very neutron-rich systems, there may exist a distribution of the excited nuclear matter into a neutron-rich gas and a more symmetric liquid. However, Lee and Mekjian [7] have pointed out that the Coulomb and surface effects, which are important for finite systems, may moderate the neutron enrichment of the gas phase by producing more free protons. The statistical multifragmentation model (SMM) calculations [8] have shown that the neutron content of the IMF can increase in the region of the phase transition, in agreement with the experimental observations [9].

In this situation only new experimental data could solve the problem of isospin composition of the gas and liquid phases. Indeed, a variety of experiments can be found in literature. In Refs. [10,11] the difference of the mean N/Z ratio of the LCP and of the IMF has been interpreted as the separation of gas and liquid phases. The authors of Ref. [12] considered the presence of neutron rich LPC and light IMF as evidence of the neutron enrichment of the gas phase. The average N/Z ratio of fragments emitted from excited nuclear systems is seen to vary with the excitation energy and the N/Z ratio of the system. In particular in the work of Ramakrishnan *et al.* [13], where for different reaction the beam energy and the mass of the system were kept constant, it has been shown that the IMF's isospin present a linear dependence on the N/Z ratio of the system. Moreover, it has been observed that the neutron content of the emitted IMF depends on the excitation energy of the (fixed size) sources [9], increasing as the excitation energy increase.

In this Rapid Communication we present experimental data extracted from two emitting sources with approximately the same N/Z ratio and excitation energy, but with different size. Since existing experimental data cover the study of the N/Z ratio of fragments as a function of the excitation energy and N/Z ratio of the emitting source, this analysis represents a complementary point to fill the experimental picture on the effects induced by the isospin on the decay process.

It is also important to note that these new data provide information about isospin of fragments produced in decay of the largest nuclear systems under investigation up to now ($A > 300$). This allows for more reliable extrapolations for the case of nuclear matter, as well as for the astrophysical applications in supernovae explosions and neutron stars.

The reaction Au+Au at 35 MeV/nucleon was studied in experiments performed at the National Superconducting K1200 Cyclotron Laboratory of the Michigan State University. Light charged particles and fragments with charge up to $Z=20$ were detected at $23°<\theta_{lab}<160°$ by the phoswich detectors of the MSU Miniball hodoscope [14]. The MULTICS array [15] covered the angular range $3°<\theta_{lab}<23°$ and allowed for a charge discrimination up to $Z=83$ and for good mass discrimination for $Z=1-6$ isotopes. The geometric acceptance of the combined array was greater than 87% of 4π.

A brief summary of the results published insofar will follow. The selection of the impact parameter $\hat{b}$ is based on the number N_c of charged particles detected [16]:

$$\hat{b}=b/b_{max}=\left(\int_{N_c}^{+\infty}P(N_c')dN_c'\right)^{1/2},$$

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

Through the analysis of energy and angular distributions of the experimental data, it was possible to identify events originating either from disassembly of a unique source formed in central collisions, or from decay of the quasiprojectile (QP) in peripheral and midperipheral collisions [17]. The excitation energy of the fragment sources was calculated both via the calorimetric method and through comparison with model calculations [17,18]. Extra kinetic energy, such as radial flow or rotational motion, does not contribute appreciably to the excitation energy. This results from the analysis of charge, angular, and kinetic energy distributions of the emitted fragments, as well as from the study of event-by-event charge partition [17–19,21]. Finally, nuclear temperatures were determined using the technique of the double ratio of isotope yields [20].

The analysis determined that the thermal characteristics (excitation energy E^* and isotope temperature T_{iso}) of the QP, formed at $0.6<\hat{b}<0.7$ ($E^*=5.5\pm0.6$ MeV/nucleon, $T_{iso}=4.3\pm0.2$ MeV), and of the unique source ($E^*=5.5\pm0.5$ MeV/nucleon, $T_{iso}=4.3\pm0.4$ MeV) are remarkably similar [17].

It is therefore important to analyze the whole picture of fragment production in these reactions. The aim of this paper is to provide new results by comparing IMF production between emitting sources of similar excitation energies ($E^*\simeq 5-6$ MeV/nucleon) and N/Z ratio ($118/79=1.49$), but of different size ($Z=79$ for the midperipheral QP, $Z=126$ for the central unique source (CUS) [18]). In particular we will focus our attention to the isospin of emitted fragments.

Charge yields of fragments obtained for the Au+Au reactions at 35 MeV/nucleon are presented in Fig. 1. The heaviest source (CUS) decays emitting fragments lighter than the QP, and in a number larger than what is expected by a simple scaling factor. In other words, the CUS undergoes a stronger disintegration at the deexcitation stage.

The different partition in the two systems affects the number of neutrons in the IMF. Figure 2 shows the isotope production yields for different Z values. The IMF emitted from the CUS are more neutron rich than those from the QP. This effect is enhanced when plotting the relative yield ratio vs the neutron excess [see Fig. 3(a)]: the ratio between the CUS and QP yields increases with (N-Z). In Fig. 3(b) we present the ratio of relative yields of neutron-rich to neutron-poor isotopes at fixed Z values, for both CUS and QP. It appears that the CUS emits preferentially the more neutron-rich fragments. The average $\langle N\rangle/Z$ value of each atomic specie versus its charge Z is presented in Fig. 4.

One can clearly see from all the figures that the disassembly of the CUS system into a relatively larger number of fragments leads to production of more neutron-rich IMF and LCP. In the previous study it was experimentally demonstrated that fixing the size of the source, but varying its excitation energy, the increase in the multiplicity of emitted fragments is accompanied to more neutron rich IMF [9]. In the present case we observe that this effect is even more pronounced.

These results are consistent with the statistical picture of disintegration of finite nuclear systems. For example, the sta-

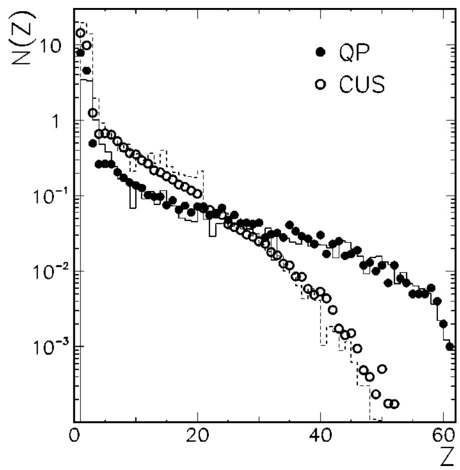

FIG. 1. Charge yields of fragments obtained for the Au+Au reactions at 35 MeV/nucleon [17,18]. Circles represent the experimental data: the solid ones are for the QP systems, the open ones are for the CUS. Solid and dashed lines are SMM calculations for peripheral and central cases, respectively.

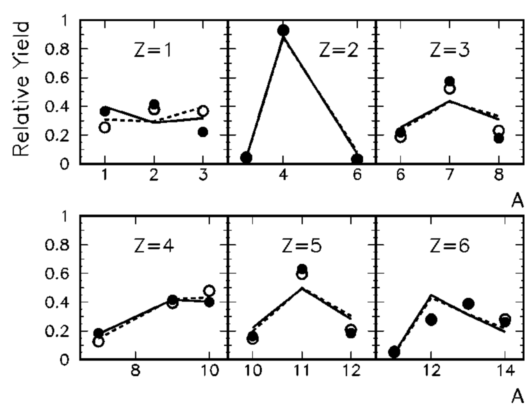

FIG. 2. Relative yields of different isotopes for fragments with charges from $Z=1$ to $Z=6$. Notations are as in Fig. 1.

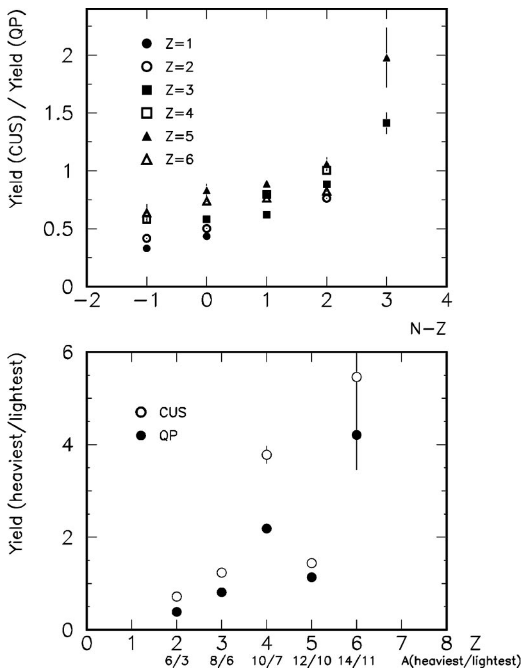

FIG. 3. (a) Ratio between the CUS and QP yields for each isotope as a function of N-Z. (b) Ratio of relative yields of neutron-rich to neutron-poor isotopes at fixed Z values, for both cases (solid points are for the QP, open points for the CUS).

tistical multifragmentation model (SMM), see, e.g., [4], is based upon the assumption of statistical equilibrium at a low-density freeze-out stage. Different breakup partitions are sampled, according to their statistical weights, in the phase space. After breakup of the nuclear source the fragments propagate independently in their mutual Coulomb field and undergo secondary decays. The deexcitation of the hot primary fragments proceeds via evaporation, fission, or Fermi-breakup [22]. SMM is very successful in reproducing experimental data concerning both peripheral and central nucleus-nucleus collisions [23–25]. In particular a very good

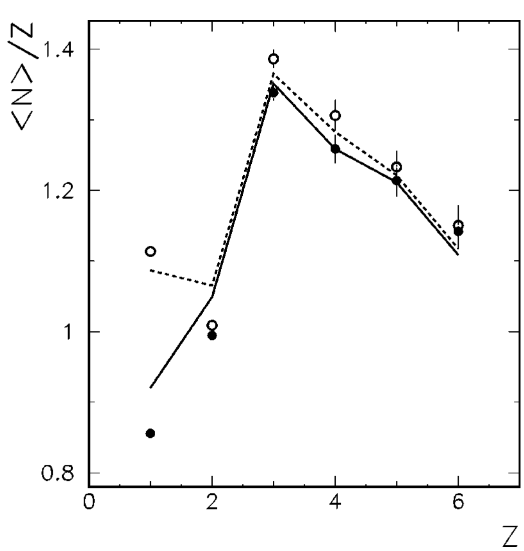

FIG. 4. Mean neutron-to-proton ($\langle N\rangle/Z$) ratio of the produced fragments. Notations are as in Fig. 1.

agreement between experimental data and SMM calculations was found in the study of the Au+Au 35 MeV/nucleon reaction [17–19]. Here we use a new version of SMM based on the generation of a Markov chain of partitions [8]. This version keeps its full reliability concerning the charge distribution predictions and allows for taking into account all effects influencing the isotope content of the produced fragments.

In this paper, by basing on this statistical model we aim to single out the main physical effects responsible for the observed trend. Even though some assumptions of the model, such as a fixed freeze-out volume or nonoverlapping fragments, are approximations of the real conditions, and some parameters (flow energy, fragment's level density) are not unambiguously defined, these uncertainties influence the fragment charge distributions mainly and can be accounted for by finding the source distribution with the well elaborated technique [21]. When the charge distributions are fixed, the main features of the studied isotope distributions are driven by the chemical equilibrium effects and the binding energy of the fragments.

The events generated by SMM were filtered to take into account the experimental efficiency. In the Figs. 1, 2, and 4 the comparisons between experimental data and SMM predictions are shown. The following set of parameters gives a good agreement between experimental data and calculations (see also [17,18,21]): an excitation energy $E_s^*=5.5\pm0.6$ MeV/nucleon; $A_s=197$, $Z_s=79$, $\rho_s=\frac{1}{3}\rho_0$, for the QP; $A_s=315$, $Z_s=126$, $\rho_s=\frac{1}{6}\rho_0$ for the CUS ($\rho_0=0.15$ fm^{-3} is the normal nuclear density). The smaller density in the central collisions is consistent with an additional expansion caused by the flow development. The microcanonical temperatures obtained in both cases are also quite similar: $T_{micr}\approx5.4$ MeV for the central case and ≈5.5 MeV for the peripheral one.

Dynamical calculations [23,26,27] predict that N/Z ratios of the emitting sources must have values close to that of the initial system. In particular an analysis dedicated to the study of 50 MeV/nucleon central collisions of^{112}Sn+ ^{112,124}Sn [27] has shown that the best agreement between calculations and experimental data claims for N/Z values of the source close to that of the starting system (differences are lower than 3%). Also the dynamical study of^{129}Xe+ ^{197}Au central and peripheral collisions at 50 MeV/nucleon has shown that the N/Z ratio of the thermal sources does not change essentially (differences are within 2%) from the initial one [26]. Results presented in Ref. [23] and concerning the study of peripheral Au+Cu reaction come to the same conclusions. Generally, since the considered emitting sources are large in size, fluctuations in their N/Z ratio value should be strongly reduced. Therefore, the assumption of conservation of the N/Z ratio after the dynamical stage seems quite realistic. We note, that our following interpretation will be valid even under less strict assumptions, namely, when the N/Z ratio changes in the same way in the both cases, or it becomes smaller for the CUS.

The reason why the nuclear sources at practically the same temperature and excitation energy produce so different fragment charge distributions (Fig. 1), can be found in the larger Coulomb energy in the CUS case, and in the different dynamics of formation of the CUS leading to a flow and, as consequence, to smaller freeze-out densities.

One can see also from the figures that the calculations reproduce the trend of increasing neutron content of produced fragments with disintegration of the nuclei into smaller pieces. This trend can be explained as follows: if the chemical equilibrium is established the big fragments have larger N/Z ratio than small ones. After disintegration of the big fragments, independently if caused by increasing excitation energy (or temperature) of a thermal source as in [9] or by a size (Coulomb) effect, the neutrons of the big fragments are accumulated mainly in the small fragments and not in free neutron gas. The SMM calculations predict that the mean number of primary free neutron in the freeze-out volume increases only from 0.98 for the peripheral source to 1.79 for the central source. This gives rise to the N/Z ratio of the IMF and LCP, which is preserved after the secondary deexcitation.

Even if the model well explains the observed effects, the experimental rise in neutron content is slightly more pronounced. One can speculate, that the initial CUS could have slightly larger N/Z ratio than the peripheral source. Our calculations show that a slight decrease of this ratio to $N/Z = 1.43$ without changing other SMM parameters, for the QP, would be sufficient to explain this disagreement. However, as we pointed to above the difference (if any) of this ratio of the sources should not be significant. For this reason, we believe that the predicted redistribution of neutrons from heavy fragments to light ones with disintegration of a nuclear system is the natural mechanism for explanation of the data.

In summary in the study of the ^{197}Au+ ^{197}Au 35 MeV/nucleon reaction it was possible to well identify two different emitting sources with the same N/Z ratio, excitation energy, and temperature ($E^* \simeq 5$–6 MeV/nucleon; $T_{iso} \simeq 4.3$ MeV from the experimental measurements, $T_{micr} \simeq 5.4$–5.5 MeV from the SMM predictions), but different size: the QP formed in midperipheral reactions and the CUS coming from the incomplete fusion of projectile and target in the most central collisions. Charge yields and isotopic content of the fragments show different distributions in the two cases. The CUS decays emitting lighter fragments than the QP and this facts reflect on the multiplicity of fragments from CUS, which is higher than expected scaling for the size factor the QP multiplicity. Thus, even if some parameters are fixed (E^*, T, N/Z) the partition of the system depends on the size and the density which determine the Coulomb energy in the freeze-out. Moreover, the neutron content of light charged particles and intermediate mass fragments increases with the size of the source.

Primary fragments produced in the freeze-out are hot and decay afterwards by emitting mainly neutrons and light charged particles. Therefore, it is natural to connect the behavior of the experimental isotope yield as a function of source size with the similar evolution of the N/Z ratio of the corresponding hot fragments. This conclusion is here supported by the SMM calculations which reproduce the fragment production in a reasonably good way. The data here presented, obtained both in central and peripheral [9] collisions, indicate that the neutron content of the fragments, produced by the decay of thermal-like systems, increases with the multiplicity of emitted IMF. The data are consistent with the hypothesis of thermal and chemical equilibration in finite nuclear systems [8], which leads to production of IMF with large neutron content. This justifies the method of extracting temperatures through isotope thermometers [17] and the thermodynamical description of these reactions.

The authors are indebted to I. N. Mishustin for his contribution to the development of the new version of SMM. The authors would also like to thank J. D. Dinius, S. Gaff, C. K. Gelbke, T. Glasmacher, M. J. Huang, W. J. Lynch, C. P. Montoya, M. B. Tsang, and H. Xi, for their collaboration during measurements and data analysis. One of the authors (A.S.B.) appreciates the warm hospitality of INFN (sezione di Trieste) where a part of this work was done.

[1] *Multifragmentation*, Proceedings of the International Workshop 27 on Gross Properties of Nuclei and Nuclear Excitations, Hirschegg, Austria, 1999 (GSI, Darmstadt, 1999).
[2] J.B. Elliot *et al.*, Phys. Rev. C **49**, 3185 (1994); M.L. Gilkes *et al.*, Phys. Rev. Lett. **73**, 1590 (1994); J.A. Hauger *et al.*, Phys. Rev. C **62**, 024616 (2000); P.F. Mastinu *et al.*, Phys. Rev. Lett. **76**, 2646 (1996); A. Bonasera, M. Bruno, C.O. Dorso, and P.F. Mastinu, Riv. Nuovo Cimento **23**, 1 (2000); M. D'Agostino *et al.*, Phys. Lett. B **473**, 219 (2000).
[3] J. Pochodzalla *et al.*, Phys. Rev. Lett. **75**, 1040 (1995).
[4] J.P. Bondorf, A.S. Botvina, A.S. Iljinov, I.N. Mishustin, and K. Sneppen, Phys. Rep. **257**, 133 (1995).
[5] D.H.E. Gross, Rep. Prog. Phys. **53**, 605 (1990).
[6] H. Müller and B.D. Serot, Phys. Rev. C **52**, 2072 (1995).
[7] S.J. Lee and A.Z. Mekjian, Phys. Rev. C **63**, 044605 (2001).
[8] A.S. Botvina and I.N. Mishustin, Phys. Rev. C **63**, 061601(R) (2001).
[9] P.M. Milazzo *et al.*, Phys. Rev. C **62**, 041602(R) (2000).
[10] M. Veselsky *et al.*, Phys. Rev. C **62**, 041605(R) (2000).
[11] E. Martin *et al.*, Phys. Rev. C **62**, 027601 (2000).
[12] H.S. Xu *et al.*, Phys. Rev. Lett. **85**, 716 (2000).
[13] E. Ramakrishnan *et al.*, Phys. Rev. C **57**, 1803 (1998).
[14] R.T. de Souza *et al.*, Nucl. Instrum. Methods Phys. Res. A **295**, 109 (1990).
[15] I. Iori *et al.*, Nucl. Instrum. Methods Phys. Res. A **325**, 458 (1993).
[16] C. Cavata *et al.*, Phys. Rev. C **42**, 1760 (1990).
[17] P.M. Milazzo *et al.*, Phys. Rev. C **58**, 953 (1998).
[18] M. D'Agostino *et al.*, Phys. Lett. B **371**, 175 (1996).
[19] M. D'Agostino *et al.*, Nucl. Phys. **A650**, 329 (1999).

[20] S. Albergo *et al.*, Nuovo Cimento A **89**, 1 (1985).
[21] P. Desesquelles *et al.*, Nucl. Phys. **A633**, 547 (1998).
[22] A.S. Botvina *et al.*, Nucl. Phys. **A475**, 663 (1987).
[23] A.S. Botvina *et al.*, Nucl. Phys. **A584**, 737 (1995).
[24] R.P. Scharenberg *et al.*, Phys. Rev. C **64**, 054602 (2001).
[25] C. Williams *et al.*, Phys. Rev. C **55**, R2132 (1997).
[26] A.S. Botvina *et al.*, Phys. At. Nucl. **58**, 1703 (1995).
[27] W.P. Tan *et al.*, Phys. Rev. C **64**, 051901(R) (2001).