Density dependence of isospin observables in spinodal decomposition

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Isotopic fluctuations in fragment formation are investigated in a quasianalytical description of the spinodal decomposition scenario. By exploiting the fluctuation-dissipation relations the covariance matrix of density fluctuations is derived as a function of the wave vector **k** for nuclear matter at given values of density, charge asymmetry, temperature, and the time that the system spends in the instability region. Then density fluctuations in ordinary space are implemented with a Fourier transform performed in a finite cubic lattice. Inside this box, domains with different density coexist, from which clusters of nucleons eventually emerge. Within our approach, the isotopic distributions are determined by the *N/Z* ratio of the leading unstable isoscalar-like modes and by isovector-like fluctuations present in the matter undergoing the spinodal decomposition. Hence the average value of the N/Z ratio of clusters and the width of the relative distribution reflect the properties of the symmetry energy. By generating a large number of events, these calculations allow a careful investigation of the cluster isotopic content as a function of the cluster density. A uniform decrease of the average charge asymmetry and of the width of the isotopic distributions with increasing density is observed. Finally, we remark that the results essentially refer to the early breakup of the system.

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I. INTRODUCTION

Reactions with charge-asymmetric systems open the possibility to learn about the properties of the symmetry term of the nuclear interaction in conditions of density and temperature away from ordinary values. In particular, the study of multifragmentation mechanisms in neutron-rich systems should allow us to get information on the behavior of the symmetry energy in density regions below saturation, where the nuclear system may undergo a liquid-gas phase transition. Constraints on the form of the density dependence of the symmetry energy are important not only for a better knowledge of the nucleon-nucleon interaction, and hence its extrapolation to the structure of exotic nuclei $[1-3]$, but also for the study of the neutron star crust and of supernova explosions, where a key issue is the clustering of low-density matter [\[4–7\]](#page-8-0).

The dynamics of first-order phase transitions is often induced by instabilities against fluctuations of the order parameter. In dissipative heavy-ion collisions, nuclear matter may be pushed inside the coexistence region of the nuclear liquid-gas phase diagram. Then, the observed abundant fragment formation may take place through a rapid amplification of spinodal instabilities. Experimental results pleading in favor of such a spinodal decomposition have recently been reported [\[8,9\]](#page-8-0). Spinodal instabilities in charge-asymmetric systems have been widely investigated from the theoretical point of view. The most important effect induced by the charge asymmetry is the so-called isospin distillation in which fragments (liquid) appear more symmetric with respect to the

initial matter, and light particles (gas) are more neutron-rich [\[10–15\]](#page-8-0). The amplitude of this effect depends on specific properties of the isovector part of the nuclear interaction, namely on the value and the derivative of the symmetry energy at low density [\[16\]](#page-8-0). Moreover, apart from the distillation effect, which determines the average fragment isotopic composition, the symmetry energy value also influences the width of the isotopic distributions. This feature has been recently exploited in the so-called isoscaling analysis [\[17\]](#page-8-0), where information on the symmetry energy behavior is extracted from the study of the ratio of the isotope yields obtained from two reactions with different charge asymmetry [\[18–21\]](#page-8-0).

In this paper we focus on a detailed study of isotopic properties of nucleon clusters, as obtained within the spinodal decomposition scenario. To select this mechanism, we consider nuclear matter initialized at a given temperature and at low density, inside a box with periodic boundary conditions, under the action of a stochastic field self-consistently determined [\[22,23\]](#page-8-0). In this case, the normal modes of the density fluctuations are plane waves. The instabilities are treated in the linear approximation (i.e., by retaining only first-order terms in the stochastic field or in the density fluctuations). This leads to a quasianalytical description of the growth of instabilities and the consequent formation of clusters of nucleons, which can be considered as excited primary fragments. For each considered system, a large variety of possible outcomes may be obtained, according to the initial density fluctuation values. In this way it is possible to collect numerous events with much reduced computational effort, allowing us to perform a thorough analysis of the isotopic content of nucleon clusters, the isospin distillation, and isotopic distributions, in connection with the ingredients of the effective nuclear interaction employed.

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We would like to stress that our analysis refers essentially to the early breakup of the system, where excited nucleon clusters, reflecting the nuclear matter properties, can be recognized. At this level, shell effects, which influence the *N/Z* ratio of real nuclei, are not considered. In particular, we will discuss the relation between the isotopic properties and the density of the primary fragments (at the moment of fragment formation). In fact, in the spinodal decomposition scenario, domains with different density values, from which clusters of nucleons eventually emerge, coexist. Clusters of intermediate mass, which may originate from the vapor phase or from the liquid phase as well, may have different isotopic properties, depending on the density value of the domain where they are formed. For instance, the isospin distillation mechanism, which is a feature of isoscalar-like oscillations, becomes more and more effective as the density gets higher in the domain considered.

In the actual disassembly of a nuclear system (in central nuclear collisions) the vapor phase is generally formed at the surface of the system, where a greater radial collective flow is also observed. Hence one could expect a relation between fragment isotopic observables and kinematical properties [\[24\]](#page-8-0). In particular, as we will discuss in the following, clusters emerging from low-density regions should be more neutronrich and with wider isotopic distributions. These features are peculiar to the spinodal decomposition scenario and to the occurrence of first-order liquid-gas transitions. Hence, in addition to the information that can be gained about the lowdensity behavior of the symmetry energy, this investigation should shed some light on the fragmentation mechanism itself.

The paper is organized as follows. In Sec. \mathbf{I} we outline the formalism developed in Refs. $[22,23]$ for infinite nuclear matter and its implementation in a finite cubic lattice. In Sec. [III](#page-4-0) we discuss the results of our calculations. Finally, in Sec. [IV](#page-7-0) a summary and conclusions are given.

II. FORMALISM

In this section we outline the main steps of the general formalism developed in Refs. [\[22,23\]](#page-8-0) to evaluate fluctuations of the one-body density for asymmetric nuclear matter inside the unstable (spinodal) region of the nuclear matter phase diagram. In our approach fluctuations of the proton and neutron densities are induced by a stochastic field that couples with the constituents of the system. By means of the fluctuationdissipation theorem the stochastic field is self-consistently determined. For nuclear matter in the spinodal region the time growth of fluctuations is essentially due to the unstable mean field. Therefore we neglect the effects of nucleon-nucleon collisions in the time evolution of fluctuations. Collisions would mainly add a damping to the growth rate of the fluctuations and should not change the main results of our calculations, at least at a qualitative level.

The approach of Refs. [\[22,23\]](#page-8-0) has reference to infinite nuclear matter, and thus normal modes are plane waves associated with wave numbers **k**. The growth of fluctuations leads to the formation of high-density domains, which can be associated with fragments. In Refs. [\[22,23\]](#page-8-0) the fragment recognition

is based on a procedure that allows one to determine the probability distribution of the domains containing correlated density fluctuations. We have then identified the pattern of correlated domains with the fragmentation pattern. In such a way, we could make predictions on the distributions of clusters. This procedure is based on an ansatz that relates the Gaussian distributions for the different modes of density fluctuations to the probability distribution of the correlated domains in the ordinary space.

In the present paper we follow a different strategy. From the probability distributions of fluctuations in **k** space, induced by the stochastic field, we directly generate a certain number of events characterized by stochastic distributions of density fluctuations in coordinate space. This is accomplished by performing a Fourier expansion in a finite cubic lattice of volume *V* .

In this way a substantial improvement of the approach of Refs. [\[22,23\]](#page-8-0) is obtained. Event-by-event analyses can be performed. Moreover, one can investigate new features of the fragmentation process, such as, for instance, the isotopic content of nucleon clusters as a function of their density.

A. Density fluctuations

The key quantity for the evaluation of the density fluctuations is the density-density response function. In a linear approximation for the stochastic field, the Fourier transform of the response function is given by the equation [\[22,23\]](#page-8-0)

$$
D_{i,j}(k,\omega) = D_i^{(0)}(k,\omega)\delta_{i,j} + \Sigma_l D_i^{(0)}(k,\omega)A_{i,l}(k)D_{l,j}(k,\omega),
$$
\n(1)

where $D_i^{(0)}(k, \omega)$ is the noninteracting particle-hole propagator and $A_{i,l}(k)$ is the Fourier transform of the nucleon-nucleon effective interaction. The subscripts of the various quantities take the values 1 and 2 for protons and neutrons, respectively. In this paper we use units such that $\hbar = c = k_B = 1$.

In asymmetric nuclear matter isovector and isoscalar fluctuations are coupled. However, one can still separate oscillations with neutrons and protons moving in phase (isoscalar-like) or out of phase (isovector-like) and add the contributions of the corresponding variances. This can be done because the time scales of isoscalar-like oscillations and of isovector-like oscillations are very different for nuclear matter with values of temperature and density sufficiently close to the borders of the spinodal region. Indeed, the growth rate of the unstable isoscalar-like modes, Γ_k , turns out to be much smaller than the real frequencies, ω_k^{iv} , of isovector-like modes [\[22,23\]](#page-8-0). Thus the two kinds of fluctuations can be considered two independent stochastic processes. In addition, for the relevant values of the magnitude of the wave vector *k*, the inequalities $\Gamma_k/T < 1$ and $\omega_k^{\text{iv}}/T > 1$ hold.

As a consequence of the linear approximation for the stochastic field, the probability distribution of density fluctuations, $P[\delta \varrho_i(\mathbf{k}, t)]$, is given by a product of Gaussian

DENSITY DEPENDENCE OF ISOSPIN OBSERVABLES IN *...* PHYSICAL REVIEW C **77**, 064606 (2008)

distributions:

$$
P[\delta \varrho_i(\mathbf{k}, t)]
$$

= $N \exp \left\{-\frac{1}{2V} \sum_{\mathbf{k}} \sum_{i,j} \delta \varrho_i^*(\mathbf{k}, t) [\sigma^2(k, t)]_{i,j}^{-1} \delta \varrho_j(\mathbf{k}, t) \right\},$ (2)

where *N* is a normalization constant. Each single factor corresponds to a stochastic process for a given wave vector **k** [\[23,25\]](#page-8-0). The covariance matrix for the isoscalar-like fluctuations,

$$
\sigma_{i,j}^2(k,t) = T C_{i,j}^{is}(k) \frac{1}{\Gamma_k} (e^{2\Gamma_k t} - 1),
$$
\n(3)

can be evaluated by taking the classical (thermodynamical) limit $\omega/T \ll 1$ of the fluctuation-dissipation relation. The coefficients $C_{i,j}^{\text{is}}(k)$ are the residues, times *i*, of the response function at the pole $\omega = i \Gamma_k$ [\[22\]](#page-8-0).

Since the time scale of the isovector-like fluctuations is much shorter than that of the growth of the unstable modes, for the covariance matrix of the isovector-like fluctuations we can take its asymptotic value for $t \to \infty$, with nuclear matter at given values of density, temperature, and asymmetry,

$$
\sigma_{i,j}^2(k) = C_{i,j}^{\text{iv}}(k),\tag{4}
$$

where $C_{i,j}^{\text{iv}}(k)$ represent the residues of the response function at the real pole ω_k^{iv} . This equation has been obtained by exploiting the fluctuation-dissipation theorem, now in the limit $\omega/T \gg 1$ [\[22\]](#page-8-0).

It should be noticed that, in the low-temperature limit, the isovector fluctuation amplitude is essentially different from the value expected at the thermodynamical limit, $\omega \ll T$, where the fluctuation variance is proportional to the temperature and inversely related to the value of the symmetry energy [\[26\]](#page-8-0).

With the aim to preserve a simple formalism, which, to some extent, allows us to perform calculations analytically, we have evaluated the response function of Eq. [\(1\)](#page-1-0) within a semiclassical approximation (Vlasov equation). However, this approximation cannot be extended to values of $k \geq k_F(k_F)$ being the neutron or proton Fermi momentum). We cure this shortcoming with an *ad hoc* specific recipe. We calculate poles and residues of the response function within the Vlasov approximation for $k \leq 1.1k_M$, where k_M is defined by $\Gamma_{k>k_M}$ < 0. With the physical parameters used in this work k_M is about the neutron Fermi momentum. For $k \geq 1.1k_M$ we take for the total variance of density fluctuation distributions its value for $k \to \infty$, $\sigma_{i,j}^2(k, t) = \delta_{i,j} \rho_i$ [\[27\]](#page-8-0), with ρ_i being the density of *i* species nucleons. This recipe is suggested by two circumstances. First, for $k_M < k < 1.1k_M$ the variance, which relaxes toward its asymptotic value since Γ_k < 0, remains appreciably larger than its limit ϱ_i for $k \to \infty$. Second, the latter is approached already for values of *k* slightly larger than $1.1k_M$, as can be shown by explicit calculations including quantum effects. The inaccurate evaluation of the response function in the interval $1.1k_M < k \le 1.3k_M$ should not introduce sizable effects, since the values of the variance in this interval are about one order of magnitude smaller than that of the most unstable mode.

B. Details of the interaction

In the present paper we adopt the same schematic Skyrmelike effective interaction as in Ref. [\[22\]](#page-8-0):

$$
\mathcal{A}_{i,j}(k) = \mathcal{A}(k) + \mathcal{S}_{i,j}(k). \tag{5}
$$

For the symmetric term $A(k)$ we use the finite-range effective interaction introduced in Ref. [\[28\]](#page-8-0):

$$
\mathcal{A}(k) = \left[A \frac{1}{\varrho_{\text{eq}}} + (\sigma + 1) \frac{B}{\varrho_{\text{eq}}^{\sigma + 1}} \varrho^{\sigma} \right] e^{-c^2 k^2 / 2}, \quad (6)
$$

where $\rho = \rho_1 + \rho_2$ is the uniform mean value of the total density of nucleons, $\varrho_{\text{eq}} = 0.16$ ^{cm^{-3}} is the density of symmetric nuclear matter at saturation, and

$$
A = -356.8
$$
 MeV, $B = 303.9$ MeV, $\sigma = \frac{1}{6}$.

The width of the Gaussian in Eq. (6) has been chosen to reproduce the surface energy term as prescribed in Ref. [\[29\]](#page-8-0).

The isospin-dependent part, $S_{i,j}(k)$, contains three different terms:

$$
S_{i,j}(k) = \frac{\partial^2 \mathcal{E}_{sym}}{\partial \varrho_i \partial \varrho_j} + \tau_i \tau_j Dk^2 + \frac{1 + \tau_i}{2} V_C(k) \delta_{i,j}, \qquad (7)
$$

with $\tau_1 = 1$ and $\tau_2 = -1$. Here \mathcal{E}_{sym} represents the potential part of the symmetry-energy density. For the coefficient of the isovector surface term we use the value $D = 40$ MeV fm³ [\[30\]](#page-8-0). Moreover, we include the Coulomb interaction $V_C(k)$ according to the approach of Ref. [\[31\]](#page-8-0). A mean-field exchange contribution

$$
V_C^{\text{ex}} = -\frac{1}{3} \left(\frac{3}{\pi}\right)^{1/3} e^2 \varrho_1^{-2/3}
$$

is also added to the bare Coulomb force.

To stress the effects of the asymmetry of the nuclear medium, we will present results obtained with two different parametrizations of the symmetry energy: one with a stronger density dependence ("superstiff" asymmetry term) and the other with a weaker density dependence ("soft" asymmetry term). In both cases the density dependence of the potential part of the symmetry-energy density can be expressed by

$$
\mathcal{E}_{sym}(\varrho_1, \varrho_2) = S(\varrho)(\varrho_2 - \varrho_1)^2, \tag{8}
$$

with

$$
S(\varrho) = \frac{2d}{\varrho_{\text{eq}}^2} \frac{\varrho}{1 + \varrho/\varrho_{\text{eq}}},\tag{9}
$$

where $d = 19$ MeV $\left[\frac{32}{10}\right]$, for the "superstiff" case, and

$$
S(\varrho) = d_1 - d_2 \varrho,\tag{10}
$$

where $d_1 = 240.9 \text{ MeV fm}^3$ and $d_2 = 819.1 \text{ MeV fm}^6$ [\[33\]](#page-8-0), for the "soft" case. It should be noticed that $S(\rho)$ is nothing but the potential part of the symmetry-energy coefficient divided by ρ , $S(\rho) = C_{sym}^{pot}(\rho)/\rho$. The inclusion of the Coulomb interaction gives rise to an overall decrease of the growth rate of density fluctuations with a corresponding contraction of the instability region in the (ϱ, T) phase diagram [\[31,34\]](#page-8-0).

C. Isospin effects

Proton and neutron densities oscillate in phase and out of phase, respectively, in the isoscalar-like fluctuations and in the isovector-like fluctuations, although with different amplitudes in general. The ratio between amplitudes is given by $\sigma_{1,1}^2(k)/\sigma_{1,2}^2(k) = \pm \sqrt{[\sigma_{1,1}^2(k)/\sigma_{2,2}^2(k)]}$, with + for the isoscalar-like case and − for the isovector-like case. This relation follows from the relevant property

$$
\det \left| C_{i,j}^{\text{is},\text{iv}}(k) \right| = 0 \tag{11}
$$

of the residues. Hence, for asymmetric matter, even in unstable isoscalar-like oscillations that lead to phase separation, protons and neutrons move with different amplitude. In particular one observes that the ratio between proton and neutron fluctuations is larger than the *Z/N* ratio of the original matter, leading to a more symmetric liquid phase, the so-called isospin distillation effect [\[10–15\]](#page-8-0). The ratio between the proton and neutron density fluctuations is mostly determined by the isoscalar-like fluctuations. This is simply related to the fact that, during the spinodal decomposition, the isoscalar-like fluctuations are much larger than the isovector-like ones. Thus, this ratio, for a given value of *k*, can be put as

$$
\frac{\delta \varrho_1(\mathbf{k})}{\delta \varrho_2(\mathbf{k})} \simeq \sqrt{\frac{C_{1,1}^{\rm is}(k)}{C_{2,2}^{\rm is}(k)}},\tag{12}
$$

where the ratio of the residues at the imaginary pole, $i\Gamma_k$, of $D_{i,j}(k,\omega)$ is given by

$$
\frac{C_{1,1}^{is}(k)}{C_{2,2}^{is}(k)} = \frac{D_1^{(0)}(k, i\Gamma_k)[1 - D_2^{(0)}(k, i\Gamma_k)A_{2,2}(k)]}{D_2^{(0)}(k, i\Gamma_k)[1 - D_1^{(0)}(k, \omega)A_{1,1}(k)]}.
$$
 (13)

The noninteracting particle-hole propagator, in the semiclassical approximation, is expressed by [\[22\]](#page-8-0)

$$
D_i^{(0)}(k, i\Gamma_k) \simeq -\frac{\partial \varrho_i}{\partial \tilde{\mu}_i} + \frac{1}{2\pi} m^2 F(\beta \tilde{\mu}_i) \frac{\Gamma_k}{k},
$$

where the effective chemical potential $\tilde{\mu}_i$ of neutrons or protons is measured with respect to the uniform mean field $U_i(\varrho_1, \varrho_2)$ of the unperturbed initial state, and $F(\beta \tilde{\mu}_i)$ is the function

$$
F(\beta\tilde{\mu}_i)=\frac{1}{e^{-\beta\tilde{\mu}_i}+1},
$$

with $\beta = 1/T$ being the inverse temperature. For the physical parameters considered in the present paper the second term in the expression of $D_i^{(0)}(k, i\Gamma_k)$ can be neglected at a satisfying approximation, then Eq. (13) becomes

$$
\frac{C_{1,1}^{is}(k)}{C_{2,2}^{is}(k)} = \frac{\frac{\partial \tilde{\mu}_2}{\partial \varrho_2} + A_{2,2}(k)}{\frac{\partial \tilde{\mu}_1}{\partial \varrho_1} + A_{1,1}(k)} = \frac{\frac{\partial^2 f}{\partial \varrho_2^2} + A_{2,2}(k)}{\frac{\partial^2 f}{\partial \varrho_1^2} + A_{1,1}(k)},
$$
(14)

where $f = f(\varrho, \varrho_3)$, with $\varrho_3 = \varrho_2 - \varrho_1$, is the sum of the kinetic and entropy terms of the free-energy density (i.e., *f* represents the free-energy density of a noninteracting twocomponent Fermi gas with effective chemical potentials $\tilde{\mu}_i$).

For a qualitative analysis we can neglect the Coulomb interaction. For small asymmetry values, we can limit ourselves to consider only the first-order term in the expansion of Eq. (14) in powers of the asymmetry $\alpha = \rho_3/\rho$. In this case the ratio of residues is given by

$$
\frac{C_{1,1}^{is}(k)}{C_{2,2}^{is}(k)} \simeq 1 + \frac{1}{\mathcal{B}(k)} \left[4 \frac{\partial}{\partial \varrho} \frac{\partial^2 f(\varrho, \varrho_3)}{\partial \varrho_3^2} \big|_{\varrho_3 = 0} + 8 \frac{\partial S(\varrho)}{\partial \varrho} \right] \varrho \alpha, \tag{15}
$$

where $B(k)$ represents the sum of the part of the interaction of Eq. [\(5\)](#page-2-0) common to both species of nucleons and of the second derivatives of the free-energy density $f(\rho, \rho_3)$:

$$
\mathcal{B}(k) = \mathcal{A}(k) + 2S(\varrho) + Dk^2 + \frac{\partial^2 f(\rho, \varrho_3)}{\partial \varrho_3^2}\big|_{\varrho_3 = 0} + \frac{\partial^2 f(\rho, 0)}{\partial \rho^2}.
$$

Equation (15) shows that for the isospin distillation effect the derivative of the coefficient of the symmetry energy with respect to the total density plays a crucial role.

With the parameters of the interactions considered here, we get, for systems moderately inside the spinodal region, a larger distillation effect with the "superstiff" interaction. However, it should be remarked that the derivative of $S(\rho)$ depends on the density. Actually at rather low densities distillation effects are expected to be stronger with the "soft" parametrization [\[35\]](#page-8-0).

The symmetry energy also plays an important role in determining isovector fluctuations, $\delta\rho_3$. These lead to fluctuations in the $(N - Z)$ content of a given density domain of mass $A = N + Z$. For finite values of the asymmetry α , it is generally not easy to single out from the isospin-dependent interaction, $S_{i,j}(k)$, a term that can play a conclusive role in determining the widths of isotopic distributions. Only for $\alpha = 0$ and, in addition, by neglecting the Coulomb interaction, are isovector and isoscalar fluctuations decoupled. In this case the fluctuations $\delta \varrho_3$ are only due to the isovector modes, and the residues at the real pole ω_k^{iv} are solely determined by the coefficient $S(\rho)$ in the expression of the symmetry energy of Eq. [\(8\)](#page-2-0). For finite values of *α*, terms containing derivatives of $S(\rho)$ contribute to the magnitude of the residues $C_{i,j}^{\text{iv}}(k)$ as well. Generally speaking, one can expect that the strength of the isovector pole increases with the coefficient $S(\rho)$. Hence, in the limit considered here, $\omega/T \gg 1$, isovector fluctuations become larger when the symmetry energy coefficient increases. It should be noticed that also for $\alpha \neq 0$ the contributions to $\delta\rho_3$ of the isoscalar-like modes still tend to cancel out.

D. Cluster recognition

We consider a finite cubic lattice of volume *V* . We assume that the density vanishes at the surface of the box. The number of nucleons is then fixed and in the box the liquid and vapor phases coexist. Moreover, finite size effects can be taken into account in this way. However, it should be remarked that the condition of vanishing density at the border of the box imposes some symmetries to the problem. In fact, in this case the number of independent oscillations is reduced by $2³$. Accordingly, the imposed symmetries also reduce fluctuation variances. This is not so important in the isoscalar-like case, since these modes are unstable and fluctuations are amplified anyway. In contrast, isovector-like

fluctuations remain quenched (by a factor of 8). However, this does not affect our conclusions about the relative comparison of the results obtained with the different parametrizations of the symmetry energy.

To extract the spatial density $\delta \varrho_i(\mathbf{r})$, we perform a Fourier expansion of the fluctuations $\delta \varrho_i(\mathbf{k})$. The Fourier components contain the product of three sine functions:

$$
\phi_{\mathbf{k}}(\mathbf{r}) = \left(\frac{2}{L}\right)^3 \sin(k_1 x) \sin(k_2 y) \sin(k_3 z),\tag{16}
$$

with $k_i = 2\pi n_i/L$ and $x = n_x b$, $y = n_y b$, $z = n_z b$ (where n_i and $n_{x,y,z}$ take positive integer values), where *L* and *b* are the lengths of the sides of the box and of the primitive cell, respectively. The coefficients of the functions $\phi_k(\mathbf{r})$ in the Fourier expansion are linear combinations of the isoscalarlike and isovector-like Fourier coefficients $\delta \varrho_i(\mathbf{k}, t)$, given by stochastic processes with the probability distribution of Eq. [\(2\)](#page-2-0) and the ratio of proton to neutron amplitudes given by $\sigma_{1,1}^2(k)/\sigma_{1,2}^2(k)$.

The size of the box will be chosen so that the box contains a number of nucleons of the same order of magnitude as in actual heavy-ion collision experiments. The adopted size of the primitive cell, V_{cell} , is such that at least one nucleon of both species is present in the cell on average. Clusters of nucleons are associated with high-density domains and will be formed by means of a coalescence algorithm, solely based on the density of neighboring cells. Adjacent cells with a value of the density above (liquid) or below (vapor) the average density are collected together. Then, we obtain domains of higher density surrounded by domains of lower density. In this way we can investigate separately the properties of the liquid phase and of the vapor phase. Moreover, we can build observables starting from event-by-event cluster distributions.

III. RESULTS

In this section we present and discuss statistical distributions of the clusters obtained by using the coalescence recipe outlined in the previous section. In particular we focus on the isospin content of the clusters coming from both the liquid phase and the vapor phase. The values chosen for the average density, $\rho = 0.4 \rho_{eq}$, and temperature, $T = 4.5 \text{ MeV}$, are in the range expected for the multifragmentation process [\[9,13\]](#page-8-0). For the time that the system spends in the instability region, we have chosen a value of $t = 80$ fm/c. This value is compatible with that obtained within the stochastic mean-field approach of Ref. [\[35\]](#page-8-0). Moreover, in this short time interval the growth of fluctuations is still limited so that a linear approximation can be considered as reasonable. For the side of the cubic lattice containing the nuclear system, we have adopted the value $L = 21$ fm, and the side of the primitive cell has a length of 3 fm. With the chosen value of density the box contains \simeq 370 nucleons.

We have performed calculations for the two parametrizations of the symmetry energy introduced earlier and for two values of the global asymmetry, $\alpha_0 = 0.1, 0.2$. We have run 4×10^3 events for each case. With the chosen values of the parameters the average numbers of nucleons in the liquid phase and in the vapor phase are \simeq 270 and \simeq 100, respectively.

FIG. 1. (Color online) Charge distributions for clusters belonging to the high-density phase for two values of the global asymmetry α_0 , calculated with the "soft" asymmetry term, circles ($\alpha_0 = 0.1$) and triangles ($\alpha_0 = 0.2$), and the "superstiff" asymmetry term, squares $(\alpha_0 = 0.1)$ and crosses $(\alpha_0 = 0.2)$.

A. Charge distributions

To assess the validity of our approach, we have evaluated the distribution of the cluster yield. In Fig. 1 we present the charge distribution of clusters belonging to the liquid phase, calculated for two values of the global asymmetry ($\alpha_0 = 0.1, 0.2$) and for the two parametrizations of the symmetry energy considered. One can see that the four curves displayed in the figure are rather similar. This is expected from the fact that isoscalar-like modes and related variances, which determine the density growth and the appearance of fragments, do not depend much on the behavior of the symmetry energy, nor on the global asymmetry of the matter considered. Moreover, the obtained charge distribution is similar to the results of full stochastic mean-field simulations $[9,13]$ of heavy-ion collisions at \simeq 30 MeV/*A*, where similar conditions of temperature and density inside the spinodal region are encountered. This indicates that fragment size is essentially determined by the properties of the most unstable normal modes, as derived in the linear approximation. The beating of these modes leads to the rather wide charge distribution. The charge distribution of Fig. 1 compares rather well also with the experimental results of Ref. [\[9\]](#page-8-0), where fragmentation of systems of similar size is investigated.

Another observable of experimental interest is the distribution of the heaviest cluster obtained in each event. The study of this observable requires, of course, an event-by-event analysis. The results are shown in Fig. [2.](#page-5-0) Also in this case we observe that the four curves corresponding to different charge asymmetries and to different parametrizations of the symmetry energy look quite similar. Moreover, our results are similar to the predictions of full simulations and to experimental data [\[9\]](#page-8-0).

B. Isotopic properties

From Figs. [3](#page-5-0) and [4](#page-5-0) we can appreciate the isospin distillation process occurring in the spinodal decomposition of asymmetric nuclear systems. The average ratio *N/Z* is plotted as a function of *Z* for the correlated density domains of the liquid phase (see Fig. [3\)](#page-5-0) and of the vapor phase (see Fig. [4\)](#page-5-0). One can nicely see that the ratio N/Z is smaller than the initial global value for the liquid, whereas the opposite holds for the vapor phase.

FIG. 2. (Color online) Distributions of the heaviest cluster for two values of the global asymmetry α_0 . Symbols are the same as in Fig. [1.](#page-4-0)

FIG. 3. (Color online) Average ratio *N/Z* as a function of *Z* for clusters belonging to the high-density (liquid) phase, calculated with the "soft" asymmetry term (circles and triangles) and the "superstiff" asymmetry term (squares and crosses). (Bottom) Global asymmetry $\alpha_0 = 0.1$. (Top) $\alpha_0 = 0.2$. The solid horizontal line is the global ratio *N/Z* for $\alpha_0 = 0.1$ and the dashed horizontal line is the global ratio N/Z for $\alpha_0 = 0.2$.

FIG. 4. (Color online) The same as in Fig. 3 but for clusters belonging to the low-density (vapor) phase.

We notice that this effect is present also for the two coexisting phases at thermodynamical equilibrium, as shown in Refs. [\[14,26\]](#page-8-0). Moreover, we observe a slight decrease of the ratio *N/Z* with *Z*. This trend is essentially related to the fact that lighter clusters originate from lower density domains where, as discussed in the following (see Sec. [III C\)](#page-6-0), isospin distillation is less effective. We notice that this behavior, which is in agreement with the results of previous calculations based on dynamical models [\[14,36\]](#page-8-0), is not observed in statistical model calculations, which exhibit an opposite trend [\[37,38\]](#page-8-0). Hence, this property of the ratio N/Z with respect to the cluster charge allows us to disentangle the predictions of different models.

In particular, Figs. 3 and 4 show that isospin distillation increases with increasing global asymmetry and is larger when the "superstiff" asymmetry term is used with respect to the "soft" case. These features are explained by Eq. [\(15\)](#page-3-0). Indeed, the induced asymmetry is proportional to the global asymmetry α_0 and, at the considered density, the derivative $\partial S(\rho)/\partial \rho$ has nearly the same magnitude but opposite sign for the two used parametrizations of the symmetry energy (positive for the "superstiff" case and negative for the "soft" case).

Now we turn our attention to the probability distribution of the asymmetry parameter for the domains belonging to the two phases. In Figs. 5 and [6](#page-6-0) we report the probability of finding a

FIG. 5. (Color online) Average distribution of asymmetry *α* for clusters belonging to the high-density (liquid) phase, calculated with the "soft" asymmetry term (circles and triangles) and the "superstiff" asymmetry term (squares and crosses). (a) Global asymmetry α_0 = 0.1. (b) $\alpha_0 = 0.2$.

FIG. 6. (Color online) The same as in Fig. [5](#page-5-0) but for clusters belonging to the low-density (vapor) phase.

cluster with asymmetry $\alpha = (N - Z)/(N + Z)$ for the liquid and vapor phases, respectively. The distribution is averaged over intermediate mass clusters $(5 < N + Z < 50)$ for the high-density phase and over light clusters $(3 < N + Z < 15)$ for the low-density phase. The shift of the maximum toward the left (right) for the liquid (vapor) phase in the "superstiff" case with respect to the "soft" case is due to the more effective distillation power of the "superstiff" interaction, as already seen. In addition we observe that distributions are wider in the "soft" case. This effect can be related to the fact that at the considered density the coefficient $S(\rho)$ of Eq. [\(8\)](#page-2-0) is larger in the "soft" case with respect to the "superstiff" case, leading to a larger value of the residues in Eq. [\(4\)](#page-2-0). Moreover, the distributions are generally broader for the vapor phase. In the following, we will show that this feature can be easily explained on the basis of the relative contributions of isoscalar-like and isovector-like oscillations, at least within our linear treatment of fluctuations. Moreover, we observe that the asymmetry distributions become narrower with increasing global asymmetry for both phases. This was observed also in Ref. [\[26\]](#page-8-0), within a thermodynamical study of spinodal decomposition. A final remark on this topic is in order. As already seen for the distillation effect, the behavior of the width of the isotopic distributions, in connection with the parametrization adopted for the symmetry energy, depends on the density value of the matter undergoing spinodal decomposition. With the present approach the widths are slightly smaller in the "superstiff" case than in the "soft" case, whereas in Ref. [\[22\]](#page-8-0) we have observed an opposite tendency. In fact, with the choice of parameters of Ref. [\[22\]](#page-8-0), the nuclear matter explored a region of higher instability (lower density) with respect to the present calculations. Then, the ratio between the amplitudes of the isoscalar-like fluctuations (which also contribute to the isotopic variance, owing to the dependence of the distillation effect, $\delta \varrho_2/\delta \varrho_1$, on the wave number *k*) and of the isovector-like fluctuations was larger than here. Furthermore, the weighting procedure of the different fluctuation modes in Ref. [\[22\]](#page-8-0) could likely underestimate the isovector-like fluctuations.

C. Density dependence of the distillation effect

Now we show that the amplitude of the isospin distillation effect, as well as the variances of the isotopic distributions, is related to the value of the density reached in the different domains associated with nucleon clusters.

Because of the beating of the several unstable modes, clusters with a given charge are not associated with a fixed density domain but may come from domains of different volume having different density (i.e., the cluster density ρ may fluctuate). We will show that this effect influences the cluster isotopic properties.

As far as the distillation effect is concerned, we notice that the N/Z ratio of a cluster can be written as

$$
N/Z = \left(\varrho_2^0 + \delta\varrho_2\right) / \left(\varrho_1^0 + \delta\varrho_1\right),
$$

where ϱ_2^0 and ϱ_1^0 are, respectively, the neutron and proton densities of the initial matter and $\delta \varrho_2$ and $\delta \varrho_1$ are the corresponding fluctuation values in the domain considered. After some algebra one obtains

$$
N/Z = (N/Z)_0 - \frac{[(N/Z)_0 - \delta \varrho_2/\delta \varrho_1] \delta \varrho_1/\varrho_1^0}{1 + \delta \varrho_1/\varrho_1^0},
$$

where $(N/Z)_0 = \rho_2^0/\rho_1^0$. We notice that, because of the distillation effect, $\delta \varrho_2 / \delta \varrho_1 < (N/Z)_0$. From this expression, one can see that the ratio N/Z of a given cluster decreases when the density ρ of the domain increases [i.e., when $\delta \rho_1$ and $\delta \varrho_2$ are larger]. In fact, higher densities are associated with larger isoscalar-like fluctuations and thus with a larger distillation. In contrast, clusters in the vapor phase, having density lower than the initial one ($\delta \varrho < 0$), show a larger value of the ratio N/Z . Moreover, the deviation of the ratio N/Z with respect to the initial value $(N/Z)_0$ increases as the ratio $\delta\rho_2/\delta\rho_1$ gets smaller (i.e., for parametrizations of the symmetry energy that lead to a larger distillation effect). In Fig. [7](#page-7-0) we show the *N/Z* ratio, averaged over all charges, as a function of the cluster density. The decreasing trend is clear, especially for the more neutron rich system, where one can also better appreciate the different effect of the two equations of state employed in the calculations.

For the variance of the isotopic distributions, we also expect a decreasing behavior with density. In fact isovector-like fluctuations, which are mainly responsible of the isotopic variance, in our linear approach are implemented according to the properties (density, asymmetry, and temperature) of the initial matter. Then, nucleon clusters with higher density

FIG. 7. (Color online) Average ratio *N/Z* as a function of the cluster density for two values of the global asymmetry α_0 , calculated with the "soft" asymmetry term, circles ($\alpha_0 = 0.1$) and triangles $(\alpha_0 = 0.2)$, and the "superstiff" asymmetry term, squares $(\alpha_0 = 0.1)$ and crosses ($\alpha_0 = 0.2$)

should have narrower isotopic distributions owing to the larger contribution of the isoscalar-like modes with respect to isovector-like fluctuations.

The probability distribution for the isovector density fluctuations $\delta \varrho_3$ in a given domain of volume *V* can be approximatively expressed as

$$
P \approx \exp\left(-\delta \varrho_3^2/2\sigma_{\rm iv}^2\right),\tag{17}
$$

where the variance σ_{iv}^2 can be evaluated by starting from the value of $\sigma_{i,j}^2(k)$ of Eq. [\(4\)](#page-2-0) and is inversely proportional to the volume $V, \sigma_{iv}^2 = g(\varrho, T) / V$, where $g(\varrho, T)$ is a function that depends on the initial conditions of the matter undergoing spinodal decomposition and on the parameters on the interaction adopted $[16]$. Then, for a cluster of volume *V*, the distribution $P(N - Z)$ can be written as

$$
P(N - Z) \approx \exp\{-[N - Z - (N_0 - Z_0)]^2/[2Vg(\varrho, T)]\}.
$$
\n(18)

Hence, the variance of the isotopic distribution should be proportional to *V*. This is confirmed by our calculations, as shown in Fig. 8, where we plot the variance of the $(N - Z)$ distribution, averaged over all clusters with volume $V = N_V V_{cell}$, as a function of N_V . From Fig. 8 one can also see that, especially in the case of the "superstiff" interaction, the variance is smaller for the neutron richer system (compare squares and crosses). Moreover, for a given volume, variances are larger in the "soft" case.

If we now consider clusters with a given mass *A*, we can easily realize that the isotopic distributions will depend on the cluster density, being broader for clusters with lower density (i.e., greater volume). Hence clusters that originate from higher density domains are expected to have an average *N/Z* ratio that is smaller than the value corresponding to the initial matter and, at the same time, are expected to show narrower isotopic distributions.

FIG. 8. (Color online) Average variance of the (*N* − *Z*) distribution for clusters of volume $V = N_V V_{cell}$ as a function of the number of the elementary cells N_V calculated for two values of the global asymmetry α_0 . Symbols are the same as in Fig. 7.

IV. CONCLUSIONS

In this paper we have considered a piece of asymmetric nuclear matter contained in a cubic box with impenetrable walls. The values of density and temperature of the system are inside the spinodal instability region of the phase diagram. We have mainly focused our attention on the isotopic distributions of clusters coming from the breakup of the system.

In our approach the density fluctuations are considered within a linear approximation, and so our results refer to nuclear matter being not deeply inside the unstable spinodal region. Such a physical situation can occur in central collisions of heavy ions at moderate energy $(\simeq 30 \text{ MeV/A})$. Moreover, we remark that our investigation concerns the distributions just after the early breakup of the nuclear system.

It has been found that, at the considered values of density and asymmetry, the global features of the disassembly, the isospin distillation effect included, are essentially determined by the unstable isoscalar-like fluctuations of the density, whereas the widths of the isotopic distributions are mostly affected by the isovector-like oscillations.

Within the model developed it is possible to recognize clusters originating from domains having different density values. Discerning clusters with density lower or higher than the initial density, we have observed a clear occurrence of the isospin distillation effect: Low-density clusters are more neutron-rich than the initial system; correspondingly the opposite happens for higher density clusters. As a general trend the average value of the *N/Z* ratio decreases with increasing cluster density. This effect is enhanced for more charge-asymmetric systems. Moreover, it has been found that the isotopic distributions of clusters belonging to the low-density phase are broader than those of clusters belonging to the high-density phase. Hence larger distillation effects are associated with narrower isotopic distributions, at least within our linear treatment of fluctuations. This feature, if it survives the secondary decay of primary fragments, could be qualitatively checked in experiments by comparing the isotopic distributions of light clusters, which should emerge from the low-density phase, and intermediate mass fragments,

most likely associated with higher density regions. However, at the present level of investigation, a careful comparison with experiments would require a reconstruction of the properties of the primary fragments, based on actual experimental data [39].

To stress the sensitivity of the results to the isovector part of the nuclear interaction, we have presented calculations obtained with two different parametrizations of the symmetry energy. For the values of charge-asymmetry considered, we have seen that isospin effects are essentially related to the values of the coefficient of the symmetry energy and of

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its derivative with respect to the density. In particular, the former quantity mainly determines the width of the isotopic distributions, whereas the latter quantity plays a crucial role for determining the strength of the isospin distillation. However, it should be remarked that the symmetry energy properties depend on the density reached by the system during the decomposition process. Therefore the effectiveness of a given parametrization of the symmetry energy for the isospin distillation depends on the actual path followed by the nuclear system in the density-temperature phase diagram.

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