PHYSICAL REVIEW C **67**, 041602(R) (2003)

A unique spinodal region in asymmetric nuclear matter

Jérôme Margueron
GANIL CEA/DSM, CNRS/IN2P3, Boîte Postale 5027, F-14076 Caen Cedex 5, France
and Istituto Nazionale di Fisica Nucleare, Sezione di Pisa, I-56100 Pisa, Italy

Philippe Chomaz
GANIL CEA/DSM, CNRS/IN2P3, Boîte Postale 5027, F-14076 Caen Cedex 5, France
(Received 8 November 2002; published 18 April 2003)

Asymmetric nuclear matter at subsaturation densities is shown to present only one type of instability. The associated order parameter is dominated by the isoscalar density and so the transition is of liquid-gas type. The instability goes in the direction of a restoration of the isospin symmetry leading to a fractionation phenomenon. These conclusions are model independent since they can be related to the general form of the asymmetry energy. They are illustrated using density functional approaches.

DOI: 10.1103/PhysRevC.67.041602 PACS number(s): 21.65.+f, 25.70.Pq, 71.10.Ay

Phase transitions are universal phenomena of matter in interaction. The coexistence regions that correspond to thermodynamically forbidden areas exhibit general features such as metastabilities or instabilities. Since strong interaction between nucleons is of van der Waals type nuclear systems are expected to present a liquid phase and a gas phase characterized by their respective densities [1]. Since nucleons can be either protons or neutrons the transition occurs in two-fluid systems, which may lead to a richer phase diagram involving up to two order parameters. It has been argued that asymmetric nuclear matter (ANM) presents different types of instabilities: a broad chemical instability region with the concentration as order parameter and a narrower domain of mechanical instability for which the total density plays the role of a second order parameter [2,3]. In this Rapid Communication we will show that spinodal instability and phase transition in ANM involves a unique order parameter, which reflects density fluctuation between the two phases. We will stress that the transition induces variations of the concentrations leading to the isospin fractionation [4] similar to the one experimentally observed [6]. These properties, and in fact all the characteristics of the instabilities, are independent of their usual classification as chemical or mechanical instabilities. These conclusions are related to general properties such as the isospin dependence of the asymmetry energy. The shape of the spinodal region and the associated instability times depend upon the model, so the observation of spinodal decomposition may provide constraints on the isospin dependence of the effective forces.

Let us consider ANM characterized by a proton and a neutron densities $\rho_i=\rho_p,\ \rho_n$. These densities can be transformed in a set of two mutually commuting charges $\rho_i=\rho_1,\ \rho_3$, where ρ_1 is the density of baryons, $\rho_1=\rho_p+\rho_n$, and ρ_3 the asymmetry density $\rho_3=\rho_n-\rho_p$. In infinite matter, the extensivity of the free energy implies that it can be reduced to a free energy density, $F(T,V,N_i)=V\mathcal{F}(T,\rho_i)$. The system is stable against separation into two phases if the free energy of a single phase is lower than the free energy in all two-phases configurations. This stability criterion implies that the free energy density is a convex function of the densities ρ_i. A local necessary condition is the positivity of the curvature matrix,

$$[\mathcal{F}_{ij}]=\left[\frac{\partial^2\mathcal{F}}{\partial\rho_i\partial\rho_j}\bigg|_T\right]\equiv\left[\frac{\partial\mu_i}{\partial\rho_j}\bigg|_T\right], \tag{1}$$

where we have introduced the chemical potentials

$$\mu_j\equiv\frac{\partial F}{\partial N_j}\bigg|_{T,V,N_{i\neq j}}=\frac{\partial\mathcal{F}}{\partial\rho_j}\bigg|_{T,\rho_{i\neq j}}.$$

In the considered two-fluids system, the $[\mathcal{F}_{ij}]$ is a 2×2 symmetric matrix, so it has two real eigenvalues $\lambda^{\pm}$ [5],

$$\lambda^{\pm}=\frac{1}{2}\left(\mathrm{Tr}[\mathcal{F}_{ij}]\pm\sqrt{\mathrm{Tr}[\mathcal{F}_{ij}]^2-4\,\mathrm{Det}[\mathcal{F}_{ij}]}\right) \tag{2}$$

associated to eigenvectors $\boldsymbol{\delta\rho}^{\pm}$ defined by $(i\neq j)$

$$\frac{\delta\rho_j^{\pm}}{\delta\rho_i^{\pm}}=\frac{\mathcal{F}_{ij}}{\lambda^{\pm}-\mathcal{F}_{jj}}=\frac{\lambda^{\pm}-\mathcal{F}_{ii}}{\mathcal{F}_{ij}}. \tag{3}$$

Eigenvectors associated with negative eigenvalue indicate the direction of the instability. It defines a local order parameter, since it is the direction along which the phase separation occurs. The eigenvalues λ define sound velocities, c, by $c^2=(1/18m)\rho_1\lambda$. In the spinodal area, the eigenvalue λ is negative, so the sound velocity c is purely imaginary and the instability time τ is given by $\tau=d/|c|$, where d is a typical size of the density fluctuation.

The requirement that the local curvature is positive is equivalent to the requirement that both the trace ($\mathrm{Tr}[\mathcal{F}_{ij}]=\lambda^{+}+\lambda^{-}$) and the determinant ($\mathrm{Det}[\mathcal{F}_{ij}]=\lambda^{+}\lambda^{-}$) are positive,

$$\mathrm{Tr}[\mathcal{F}_{ij}]\geqslant0 \quad\text{and}\quad \mathrm{Det}[\mathcal{F}_{ij}]\geqslant0. \tag{4}$$

The use of the trace and the determinant, which are two basis-independent characteristics of the curvature matrix, clearly stresses the fact that the stability analysis should be independent of the arbitrary choice of the thermodynamical quantities used to label the state, e.g., $(\rho_p,\ \rho_n)$ or $(\rho_1,\ \rho_3)$. If Eq. (4) is violated, the system is in the unstable region of a phase transition. Two cases are then possible: (i) only one eigenvalue is negative and one order parameter is sufficient to describe the transition or (ii) both eigenvalues are negative and two independent order parameters should be considered meaning that more than two phases can coexist.

For ANM below saturation density, case (ii) never occurs since the asymmetry energy has always positive curvature ($\mathcal{F}_{33}$). Indeed, the asymmetry term in the mass formula behaves like $(N-Z)^2$ times a positive function of A showing that the dominant ρ_3 dependence of the asymmetry potential energy is essentially quadratic and that $\mathcal{F}_{33}$ is a positive function of the total density. Recent Bruckner calculations in ANM [7] have confirmed the positivity of $\mathcal{F}_{33}$. They have parametrized the potential energy [8] with the simple form $\mathcal{V}(\rho_1,\rho_3)=\mathcal{V}_0(\rho_1)\rho_1^2+\mathcal{V}_1(\rho_1)\rho_3^2$ with $\mathcal{V}_0/\mathcal{V}_1 \sim -3$ and $\mathcal{V}_0<0$. This is also true for effective forces such as Skyrme forces. For example, the simplest interaction with a constant attraction t_0 and a repulsive part $t_3\rho_1$ leads to $\mathcal{V}(\rho_1,\rho_3)=(3\rho_1^2-\rho_3^2)\beta(\rho_1)$ with $\beta(\rho_1)=(t_0+t_3/6\rho_1)/8$. The function $\beta(\rho_1)$ is negative below saturation density, hence the contribution of the interaction to $\mathcal{F}_{33}$ in the low density region is always positive.

These arguments show that, below saturation density, the ρ_3 curvature, $\mathcal{F}_{33}$, is expected to be positive for all asymmetries. Since the curvature in any direction, $\mathcal{F}_{ii}$, should be between the two eigenvalues $\lambda^-\leqslant\mathcal{F}_{ii}\leqslant\lambda^+$ we immediately see that if $\mathcal{F}_{33}$ is positive one eigen curvature at least should remain positive. In fact for all models we have studied $\mathcal{F}_{33}$ appears to be always large enough so that the trace is always positive demonstrating that $\lambda^+>0$. Since $\mathrm{Tr}[\mathcal{F}_{ij}]=\mathcal{F}_{nn}+\mathcal{F}_{pp}$, this can be related to the positivity of the Landau parameter $\mathcal{F}_{nn}$ and $\mathcal{F}_{pp}$.

The large positive value of $\mathcal{F}_{33}$ also indicates that the instability should remain far from the ρ_3 direction, i.e., it should involve total density variation and indeed we will see that in all models and for all asymmetries the instability direction hardly deviates from a constant asymmetry direction ($\delta\rho_3\ll\delta\rho_1$). This isoscalar nature of the instability can be understood by looking at the expression of the eigenmodes in the (ρ_n,ρ_p) coordinates. Since $\lambda^-\leqslant\mathcal{F}_{ii}\leqslant\lambda^+$, the differences $\lambda^--\mathcal{F}_{ii}$ are always negative demonstrating, using Eq. (3), that the instability is of isoscalar type, if the Landau parameter $\mathcal{F}_{np}$ is negative. Hence, there is a close link between the isoscalar nature of the instability and the attraction of the proton-neutron interaction [5].

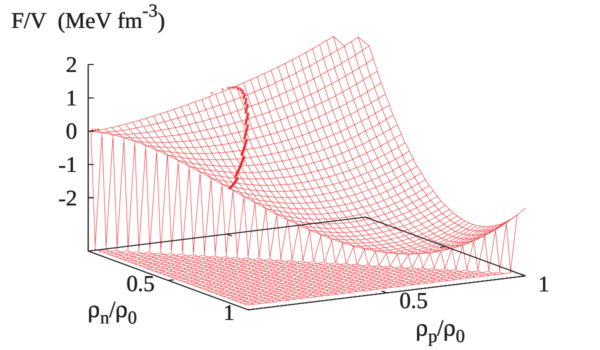

FIG. 1. This figure represents the energy surface as a function of the densities ρ_n and ρ_p for the SLy230a interaction. The contours delimitate the spinodal area.

To illustrate the above results we will now use density functional formalism using Skyrme [9] and Gogny [10] effective forces. It should be noted that the extraction of the sound velocity corresponds to a random phase approximation (RPA), which goes beyond the mean field approximation. We will focus on the isospin degree of freedom for which RPA approaches can be considered as good approximations and discuss both zero and finite temperatures in ANM.

We represent in Fig. 1 the energy surface as a function of ρ_n and ρ_p, deduced from the SLy230a Skyrme interaction [11]. In the symmetric case ($\rho_n=\rho_p$), one can see the negative curvature of the energy which defines the spinodal area, whereas in pure neutron matter ($\rho_p=0$), no negative curvature and so no spinodal instability are predicted. We can also notice that the isovector density dependence is almost parabolic illustrating the positivity of $\mathcal{F}_{33}$.

The spinodal contours predicted by several models exhibit important differences (see Fig. 2). In the case [14] of SLy230a force (as well as SGII, D1P), the total density at which spinodal instability appears decreases when the asymmetry increases, whereas for SIII (as well as D1, D1S) it increases up to large asymmetry and finally decreases. Considering the implicit equation of the spinodal limit

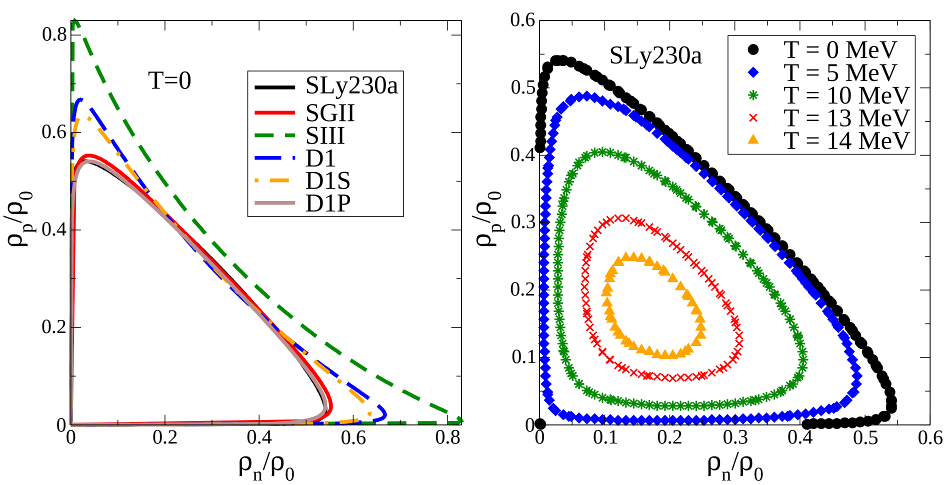

FIG. 2. These two figures are a projection of the spinodal contour in the density plane: left, for Skyrme (SLy230a [11], SGII [12], SIII [13]) and Gogny models (D1 [10], D1S [15], D1P [16]); right, temperature dependence of the spinodal zone computed for the SLy230a case.

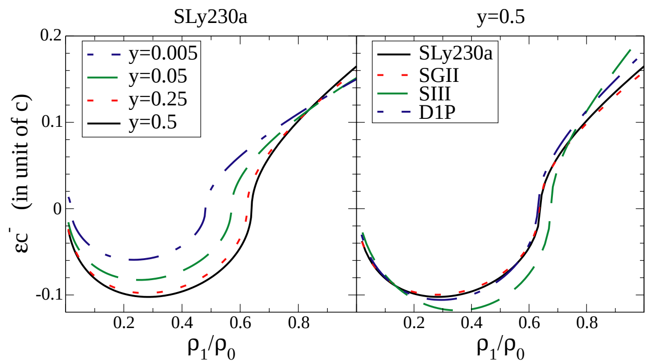

FIG. 3. Sound velocity as a function of the density. Negative values of sound velocity mean that it is imaginary ($\epsilon = i$ is c^- imaginary). On the left, we have changed the asymmetry parameter and fixed the model (SLy230a), on the right, we have changed the models and fixed $y = \rho_p/\rho_1 = 0.5$.

$g(\rho_1,\rho_3) \equiv \mathrm{Det}[\mathcal{F}_{ij}] = 0$, we can show that the curvature of the spinodal around the symmetry is related to $\partial^2 g/\partial\rho_3^2(\rho_3 = 0)$. The isospin symmetry imposes that $\partial g/\partial\rho_3(\rho_3 = 0) = 0$ and one can show that

$$\left.\frac{\partial^2 g}{\partial\rho_3^2}\right|_{\rho_3=0} = \mathrm{Det}\left[\left.\frac{\partial\mathcal{F}_{ij}}{\partial\rho_3}\right|_{\rho_3=0}\right] + (\mathcal{F}_{1133}\mathcal{F}_{33} + \mathcal{F}_{3333}\mathcal{F}_{11} + \mathcal{F}_{1333}\mathcal{F}_{13}). \qquad (5)$$

Assuming that the fourth derivatives of the free energy are negligible compared to the third one, the curvature of g is mainly given by $\mathrm{Det}[\partial\mathcal{F}_{ij}/\partial\rho_3]$, where $\partial\mathcal{F}_{ij}/\partial\rho_3$ is nothing but the curvature matrix of $\partial F/\partial\rho_3$, i.e., the asymmetry density dependence of the free energy. We observe that all forces which fulfill the global requirement that they reproduce symmetric nuclear matter (SNM) equation of state as well as the pure neutron matter calculations, leads to the same curvature of the spinodal region.

The temperature dependence of the spinodal contour can be appreciated in the right panel of Fig. 2. As the temperature increases, the spinodal region shrinks up to the critical temperature for which it is reduced to the SNM critical point. However, up to a rather high temperature (5–10 MeV) the spinodal zone remains almost identical to the zero temperature one.

We show in the right panel of Fig. 3 the sound velocity in SNM as a function of the density for several forces. When we enter into the spinodal area, the sound velocity becomes purely imaginary. The different forces predict different instability time. However, for the set of forces fitted to reproduce the symmetric and the pure neutron matter calculation, we observe a convergence of predictions as we already observed on the spinodal boundaries. In the left panel of Fig. 3 we can appreciate the reduction of the instability when we go away from SNM. However, large asymmetries are needed to induce a sizable effect.

Various contours of equal imaginary sound velocity are represented in Fig. 4 for SLy230b and D1P interactions. The more internal curves correspond to the sound velocity $i0.09c$, after comes $i0.06c$, $i0.03c$, and finally 0, the spinodal border. For these two recent forces that take into account pure neutron matter constraints, the predicted instability domains are rather similar. We observe that in almost all the spinodal regions, the sound velocity is larger than $0.06c$.

Let us now focus on the direction of the instability. If $\boldsymbol{\delta\rho}^-$ is along $y = \rho_p/\rho_1 = \mathrm{const}$ then the instability does not change the proton fraction. For symmetry reasons, pure isoscalar ($\delta\rho_3 = 0$) and isovector ($\delta\rho_1 = 0$) modes appear only for SNM. So it is interesting to introduce a generalization of isoscalarlike and isovectorlike modes, by considering if the protons and neutrons move in phase ($\delta\rho_n^- \delta\rho_p^- > 0$) or out of phase ($\delta\rho_n^- \delta\rho_p^- < 0$). Figure 4 shows the direction of instabilities along the spinodal border and some isoinstability lines. We observed that the instability is always almost along the ρ_1 axis, meaning that it is dominated by total density fluctuations even for large asymmetries. Figure 5 presents the angle of the eigenstate $\boldsymbol{\delta\rho}^-$ with the isoscalar axis normalized by the angle between the $y = \mathrm{const}$ line and the isoscalar axis (denoted χ) for three models (D1P, SGII, SLy230a). We can see that this quantity is in between 0 and 1, so that the instability direction is between the $y = \mathrm{const}$

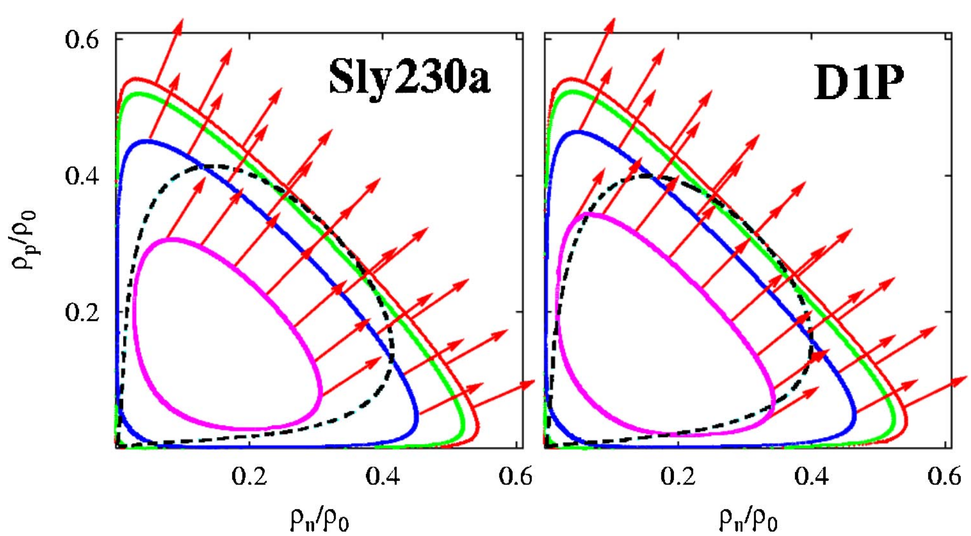

FIG. 4. This is the projection of the iso-eigenvalues on the density plane for Slya (left) and D1P (right). The arrows indicate the direction of instability. The mechanical instability is also indicated (dotted line).

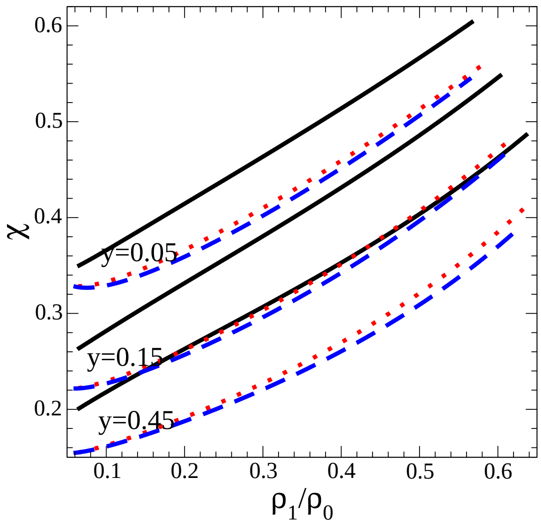

FIG. 5. Angle of $\boldsymbol{\delta\rho}^-$ with the isoscalar axis normalized by the angle between the $y=$const line and the isoscalar axis (denoted χ) for various values of y noted on the figure and for three models (solid: SLy230a, dotted: D1P, dashed: SGII).

line and the ρ_1 direction. This shows that the unstable direction is of isoscalar nature, as expected from the attractive interaction between proton-neutron. The total density is, therefore, the dominant contribution to the order parameter showing that the transition is between two phases having different densities (i.e., liquid-gas phase transition). The angle with the ρ_1 axis is almost constant along a constant y line. This means that, as the matter enters in the spinodal zone and then dives into it, there are no dramatic changes in the instability direction, which remains essentially a density fluctuation. Moreover, the unstable eigenvector drives the dense phase (i.e., the liquid) towards a more symmetric point in the density plane. By particle conservation, the gas phase will be more asymmetric leading to the fractionation phenomenon. Those results are in agreement with recent calculations for ANM [5] and nuclei [17].

We want to stress that those qualitative conclusions are very robust and have been reached for all the Skyrme and Gogny forces we have tested (SGII, SkM*, RATP, D1, D1S, D1P, etc.) including the most recent ones (SLy230a, D1P) as well as the original ones (such as SIII, D1).

A different discussion can be found in the literature [2,3]. Therein, chemical and mechanical stability conditions

$$\left.\frac{\partial\mu_p}{\partial y}\right|_{T,P}>0 \quad\text{and}\quad \left.\frac{\partial P}{\partial\rho_1}\right|_{T,y}>0 \tag{6}$$

are introduced and two types of spinodal regions are defined. This leads to the idea that two order parameters should be introduced: the concentration in the chemical instability zone and the baryon density in the mechanical instability region. To try to connect the eigenstate analysis with this discussion, one can use the relations

$$\rho_n \mathrm{Det}[\mathcal{F}_{ij}]=\left.\frac{\partial\mu_p}{\partial y}\right|_{T,P}\left.\frac{\partial P}{\partial\rho_1}\right|_{T,y}. \tag{7}$$

Comparing expression (7), with the identity $\mathrm{Det}[\mathcal{F}_{ij}]=\lambda^+\lambda^-$ one can be tempted to relate separately

$$\left.\frac{\partial\mu_p}{\partial y}\right|_{T,P} \quad\text{and}\quad \left.\frac{\partial P}{\partial\rho_1}\right|_{T,y}$$

to the two eigenvalues λ^+ and λ^-. This is indeed correct in SNM [5] but these relations break down in ANM. For instance,

$$\left.\frac{\partial P}{\partial\rho_1}\right|_{T,y}=\rho_1\left.\frac{\partial^2\mathcal{F}}{\partial\rho_1^2}\right|_{T,y}$$

is nothing but the curvature of the free energy in the particular direction of constant proton fraction and this direction has no reason to be an eigenvector direction, except in SNM. Considering Eq. (7) only the product

$$\left.\frac{\partial\mu_p}{\partial y}\right|_{T,P}\times\left.\frac{\partial P}{\partial\rho_1}\right|_{T,y}$$

should be used to spot the instability region. Equation (7) shows that at the onset of instability where $\mathrm{Det}[\mathcal{F}_{ij}]=0$ the product should vanish. On the spinodal border, $(\partial P/\partial\rho_1)|_{T,y}$ vanishes only if the direction of instability is the $y=$const line. This happens only for SNM, therefore in general $(\partial\mu_p/\partial y)|_{T,P}$ should vanish first irrespectively of the actual nature of the instability. In particular, the previous eigenvector analysis shows that in the so-called chemical region, not only the concentration fluctuations δy are amplified but mainly the total density one, $\delta\rho_1$. When entering in the mechanical spinodal zone (shown in Fig. 4) nothing special happens, the instability strength evolves smoothly (see also Fig. 3) and the associated vector keeps pointing in the same isoscalarlike direction (see Fig. 5). Consequently, the chemical and mechanical stability conditions should not be considered separately but combined into the determinant of the curvature matrix.

In this Rapid Communication, we have shown that ANM does not present two types of spinodal instabilities, mechanical and chemical, but only one that is dominantly of isoscalar nature as a consequence of the negativity of the Landau parameter $\mathcal{F}_{np}$. This general property can be linked to the positivity of the symmetry energy curvature $\mathcal{F}_{33}$. This means that the instability is always dominated by density fluctuations and so can be interpreted as a liquid-gas separation. The instabilities tend to restore the isospin symmetry for the dense phase (liquid) leading to the fractionation of ANM. We have shown that changing the asymmetry up to $\rho_p<3\rho_n$ does not change quantitatively the density at which instability appears, nor the imaginary sound velocity compared to those obtained in SNM. All the above results are not qualitatively modified by the temperature, which mainly introduces a reduction of the spinodal region up to the SNM critical point where it vanishes. The quantitative predictions concerning the shape of the spinodal zone as well as the instability times depend upon the chosen interaction but converge for the various forces already constrained to reproduce the pure neutron matter calculation.

We want to thank Bao-An Li and V. Baran for interesting discussions during the preparation of this manuscript.

[1] G. Bertsch and P.J. Siemens, Phys. Lett. **126B**, 9 (1983).
[2] H. Muller and B.D. Serot, Phys. Rev. C **52**, 2072 (1995).
[3] Bao-An Li and M.C. Ko, Nucl. Phys. **A618**, 498 (1997).
[4] V. Baran *et al.*, Nucl. Phys. **A632**, 287 (1998).
[5] V. Baran *et al.*, Phys. Rev. Lett. **86**, 4492 (2001).
[6] H.S. Xu *et al.*, Phys. Rev. Lett. **85**, 716 (2000).
[7] I. Vidaña and I. Bombaci, Phys. Rev. C **66**, 045801 (2002).
[8] J. Margueron, Ph.D. thesis, IPN Orsay, 2001.
[9] T.H.R. Skyrme, Philos. Mag. **1**, 1043 (1956).
[10] D. Gogny, in *Proceedings of the International Conference on Nuclear Self-Consistent Fields*, edited by Trieste, G. Ripka, and M. Porneuf (North Holland, Amsterdam, 1975).
[11] E. Chabanat *et al.*, Nucl. Phys. **A627**, 710 (1997).
[12] Nguyen Van Giai and H. Sagawa, Nucl. Phys. **A371**, 1 (1981).
[13] M. Beiner *et al.*, Nucl. Phys. **A238**, 29 (1975).
[14] D. Gogny and R. Padjen, Nucl. Phys. **A293**, 365 (1977).
[15] J.-F. Berger *et al.*, Comput. Phys. Commun. **63**, 365 (1991).
[16] M. Farine *et al.*, J. Phys. G **25**, 863 (1999).
[17] M. Colonna *et al.*, Phys. Rev. Lett. **88**, 122701 (2002).