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Liquid-gas phase transition and instabilities in two-component systems with asymmetric interaction

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The liquid-gas phase transition and associated instability in two component systems are investigated using a mean field theory. The importance of the roles of both the Coulomb force and the symmetry energy terms are studied. The addition of the Coulomb terms brings asymmetry into a mean field interaction and thus results in important differences with previous approaches, which did not include such terms. The asymmetric Coulomb effects modify the chemical instability and mechanical instability domains shifting many features, such as the line of equal concentration and the contact line of chemical and mechanical instability boundary, away from proton fraction point $y=1/2$ to a value closer to the valley of β stability. Thus, isospin fractionization is somewhat moderated by the presence of the Coulomb force. These features and characteristics of phase transition away from β stability are discussed in detail.

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Thermodynamic properties of two and, more generally, multicomponent systems are of interest in many areas of physics. For example, binary systems of two components are encountered in nuclei where the two components are neutrons and protons. In fact, nuclei offer an interesting situation where volume, surface, symmetry energy, and Coulombic asymmetric terms are simultaneously present. While light nuclei have $N=Z$, heavy nuclei have $N>Z$, and systems with large neutron excess such as neutron stars exist in astrophysics. In condensed matter physics, two-component systems appear in liquid ^{3}He where the two-components are spin-up and spin-down fluids. Other two-component systems are binary alloys. Such two-component systems contain a more complex phase structure than one-component systems. This paper is an extension of previous work concerning the nuclear case, which was discussed from both the fragmentation point of view [1–3] and a mean field picture, [4]. In the mean field picture we studied the phase surface associated with the liquid-gas phase transition incorporating the role of surface and Coulomb effects [4]. Previous mean studies [5] of two-component systems were investigations of the role of the symmetry energy on the phase structure of the liquid-gas phase transition. While the symmetry energy plays an important role in governing the proton/neutron ratios in the liquid and gas phases, favoring more symmetric systems in the liquid phase than the gas phase (isospin fractionization), the effects of both the Coulomb interaction and the surface energy were also shown to be important. This was first discussed in Refs. [2,4] and more recently in Ref. [6]. Here, we will focus on the stability, metastability, and instability of asymmetric systems of protons and neutrons. Such systems are of interest for radioactive beams studies that can be used to explore nuclei at the limits of isospin asymmetry. In a recent paper, Baran *et al.* [7] have pointed out some new effects associated with two-component systems where a new kind of liquid-gas phase transition associated with chemical instability may appear. This new type of transition may manifest itself as an isospin distillation or fractionization as reported in Refs. [8,9] where the gas phase is enriched in neutrons relative to the liquid phase. Such observations suggest that neutron diffusion occurs at the same rate or is more rapid than fragment production. These observations are based solely on symmetry energy effects, which make the liquid phase more symmetric, with the role of the Coulomb interaction not included. Extensions by Colonna *et al.* [10] to finite nuclei show that Coulomb and surface effects reduce instability regions. A similar reduction was previously found with respect to the binodal surface by the present authors [4], which showed that the Coulomb force reduces the binodal surface to smaller one while the surface effect shifts the binodal surface down to lower pressure. The results of Colonna *et al.* also show that octupolar modes are very important and dominate the development of the instability. The importance of Coulomb effects driven by Z^2 terms versus symmetry energy effects arising from $(N-Z)^2$ terms can be seen by just looking at the most stable nucleus for a given A. Using a liquid drop model, the Z with the smallest mass or greatest binding energy (β stability) is given by $Z=(A/2)/(1+B_cA^{2/3}/4B_s)$, where B_c is the Coulomb coefficient $B_cZ^2/A^{1/3}$ and B_s is the symmetry energy coefficient $B_s(N-Z)^2/A$. When the Coulomb interaction is turned off, Z is simply $A/2$. Moreover, the Coulomb interaction was shown [2] to have a very dramatic effect on the specific heat associated with a first-order liquid-gas phase transition, substantially reducing the peak previously reported in Ref. [3], where surface energy effects associated with one-component systems played the dominant role. Furthermore, the Coulomb force introduced another pair of coexistence densities having larger proton concentration in the gas phase than in the liquid phase due to its asymmetry in the mean field interaction [4]. We study, in this paper, the effects of Coulombic asymmetric interaction to the chemical and mechanical instability for two-component system with the interaction energy density of the form

$$U(\rho)=-a_0\rho^2+a_3\rho^3+a_S(2y-1)^2\rho^2+Cy^2\rho^2 \qquad (1)$$

with y the proton fraction. The third term is the symmetry energy which is zero for symmetric systems ($y=1/2$) and the last term is the asymmetric Coulomb interaction.

In symmetric systems, mechanical instability from $dP/d\rho<0$ and chemical instability from $d\mu/d\rho<0$ at a fixed T are simply related since

$$Nd\mu=-SdT+VdP, \qquad (2)$$

where μ is the chemical potential, S is the entropy, P is the pressure, N is the number of particles, and T is the temperature. Thus, $\rho d\mu/d\rho=dP/d\rho$ at a fixed T and the boundaries of both instabilities are common with $d\mu/d\rho=dP/d\rho=0$. The coexistence curve of the Maxwell pressure P versus density ρ or volume V is determined by the equality of the pressure and chemical potentials between the liquid and the gas phases at the same T. In the region between the spinoidal curve determined by the instability line and the coexistence line, there are metastable regions of supercooled liquid and superheated gas where nucleation processes can occur [11–13].

In two-component systems, the coexistence line of a one-component system now becomes a surface in (P,y,T), with y the proton fraction. This surface has several new elements associated with it, which are not present in one-component systems. For example, the isolated critical point of a one-component system now becomes a line of critical points with $y=1/2$ the critical point of a one-component system. Associated with this surface is also a line of maximal asymmetry in the proton/neutron ratio and a line of equal concentration $y_E(T)$ in liquid and gas phases [4,5]. The intersection of this (P,y,T) with a plane at a fixed T leads to loops of P versus y as shown in Fig. 4 of Ref. [4]. The chemical potentials μ_n and μ_p versus y at fixed T and P also have interesting behavior as shown in Fig. 3 of Ref. [4]. For a two-component system, Eq. (2) is replaced by

$$y(d\mu_p/d\rho)+(1-y)(d\mu_n/d\rho)=(1/\rho)(dP/d\rho),$$

$$y(d\mu_p/dy)+(1-y)(d\mu_n/dy)=(1/\rho)(dP/dy).$$

In first equation, y and T are held fixed, and in second equation ρ and T are held fixed. We will use these figures to study the spinoidal instability region and its associated surface in (P,y,T).

Some properties of the instability region can best be analyzed using some thermodynamic identities. We first note that the pressure and the chemical potentials are usually written as functions of ρ, y, and T; $P(\rho,y,T)$ and $\mu_q(\rho,y,T)$. One important identity arises from the equation of state $P=P(\rho,y,T)$, which gives

$$\left(\frac{dy}{d\rho}\right)_{P,T}=-\left(\frac{dP}{d\rho}\right)_{y,T}\Bigg/\left(\frac{dP}{dy}\right)_{\rho,T} \qquad (3)$$

for the relation of y and ρ at fixed P and T. Since P, T, y, and ρ are connected by an equation of state $P(\rho,y,T)$, we can write

$$\left(\frac{d\mu_q}{dy}\right)_{P,T}=\left(\frac{d\mu_q}{dy}\right)_{\rho,T}+\left(\frac{d\mu_q}{d\rho}\right)_{y,T}\left(\frac{d\rho}{dy}\right)_{P,T}. \qquad (4)$$

Combining Eqs. (3) and (4), we can write $(dP/d\rho)_{y,T}$ as

$$\left(\frac{dP}{d\rho}\right)_{y,T}=\frac{-(dP/dy)_{\rho,T}(d\mu_q/d\rho)_{y,T}}{(d\mu_q/dy)_{P,T}-(d\mu_q/dy)_{\rho,T}}. \qquad (5)$$

Since $(d\mu_q/dy)_{\rho,T}$ is finite, singularities in $(d\mu_q/dy)_{P,T}$ (chemical instability) give rise to zeroes in $(dP/d\rho)_{y,T}$. The zeroes of $(dP/d\rho)_{y,T}$ form the spinodal surface, boundary for mechanical instability region. In two-component systems, mechanical and chemical instability regions are separated for asymmetric cases. Using Eq. (3), the chemical instability boundary $[(d\mu_q/dy)_{P,T}=0]$, determined by setting Eq. (4) equal to 0, is obtained from

$$\left(\frac{dP}{d\rho}\right)_{y,T}=\left(\frac{dP}{dy}\right)_{\rho,T}\frac{(d\mu_q/d\rho)_{y,T}}{(d\mu_q/dy)_{\rho,T}}. \qquad (6)$$

The following features should be noted. First, mechanical equilibrium $[(dP/d\rho)_{y,T}=0]$ is determined by the condition that the left-hand side of this equation is 0. Thus, the mechanical and chemical instability boundaries are the same when $(dP/dy)_{\rho,T}=0$ since $(d\mu_q/dy)_{\rho,T}\neq 0$ here. Due to Eq. (3), $(d\rho/dy)_{P,T}=0$ too when $(dP/dy)_{\rho,T}=0$ except on the mechanical instability boundary. This condition is also related to the equal concentration y_E of liquid and gas phases on the coexistence surface [4]. For systems without Coulomb terms, or more generally, any asymmetric term which favors one-component over another, this occurs at $y_E=1/2$. Second, for chemical equilibrium, the right-hand side can be obtained from the equation of state $P=P(\rho,y,T)$ and the behavior of the proton or neutron chemical potentials $\mu_q=\mu_q(\rho,y,T)$. q will distinguish one-component from the other, and, in particular, the neutron results from the proton results for the nuclear case. We will use Eq. (6) to explore the boundary of chemical equilibrium.

To keep equations simple in the discussions of various new features in the two-component system with the asymmetric interaction energy given by Eq. (1), we will drop the degeneracy corrections. However, these are included in the actual calculations done and in the figures (Figs. 1 and 2) presented here. In this limit, $P=\rho T-a_0(y)\rho^2+2a_3\rho^3+Cy^2\rho^2$ where we have followed the notation of Ref. [11] with P now generalized to the case of asymmetric systems. The generalization changes a_0 to $a_0(y)=a_0[1-(\frac{2}{3})(\frac{1}{2}+x_0)(2y-1)^2]$ with $a_0=-(\frac{3}{8})t_0$ and $a_3=t_3/16$. Then, $(dP/d\rho)_{y,T}=0$ at

$$\rho_\pm=(a_0(y)-Cy^2\pm\sqrt{(a_0(y)-Cy^2)^2-6a_3T})/(6a_3).$$

When $(d^2P/d\rho^2)_{y,T}=0$, the two roots are equal at $T_c^P(y)=[a_0(y)-Cy^2]^2/(6a_3)$ and are $\rho_c^P(y)=\rho_+=\rho_-=(a_0(y)-Cy^2)/(6a_3)$. At this point, the pressure is $P_c(y)=(a_0(y)-Cy^2)^3/(108a_3^2)$. Under the same limits, the chemical potentials are given by

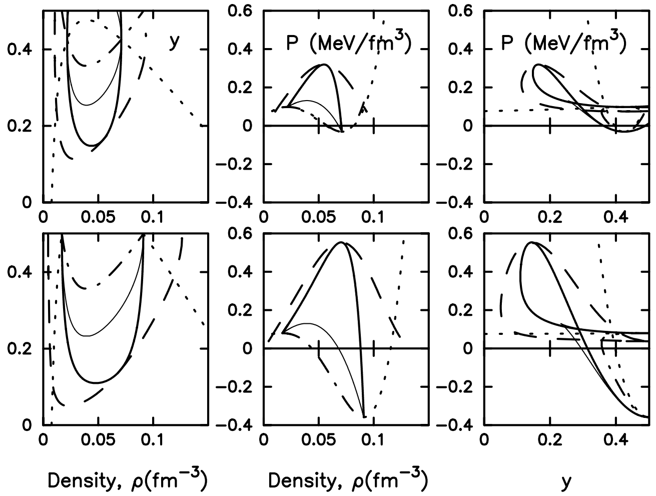

FIG. 1. Chemical (thick solid curve) and mechanical (thin solid curve) instability curves at temperature $T=10$ MeV with (upper boxes) and without (lower boxes) Coulomb effects arising from a $R=8$ fm uniform sphere. The dashed curves are the coexistence curve at $T=10$ MeV. The dotted curves are for $(d\mu_p/d\rho)_{y,T}=0$ and the dash-dotted curves are for $(d\mu_n/d\rho)_{y,T}=0$. The equal concentration $y_E=0.4256$ at $T=10$ MeV. The curves are the constant T plane cuts of the corresponding surfaces in (ρ,y,T) space (left column), in (ρ,P,T) space (center column), and in (P,y,T) space (right column).

$$\mu_q=T\ln[(\lambda^3/\gamma)(\rho/2\pm(2y-1)\rho/2)]-2a_0\rho+3a_3\rho^2$$
$$\pm(4/3)(1/2+x_0)a_0(2y-1)\rho+(1\pm1)Cy\rho$$

with the upper sign for protons and the lower sign for neutrons and $\gamma=2$ for the spin-up and spin-down degeneracies. This equation for $\mu_q=\mu_q(\rho,y,T)$ when combined with the equation of state $P(\rho,y,T)$ can be used to calculate the chemical potentials as a function $\mu_q=\mu_q(P,y,T)$. Boundaries of the chemical instability region can also be determined using the condition $d\mu_q/dy=0$ at constant T and P. Substituting the simplified equation of state and expression for the chemical potential into Eq. (6) leads to a cubic equation $c_3\rho^3+c_2\rho^2+c_1\rho+c_0=0$ with $c_3=[32(\frac{1}{2}+x_0)a_0+12C]a_3y(1-y)$, $c_2=6a_3T-[(\frac{32}{3})(\frac{1}{2}+x_0)a_0^2+(\frac{8}{3})(1-x_0)a_0C]y(1-y)$, $c_1=[-(\frac{4}{3})(1-x_0)a_0+2Cy]T$, and $c_0=T^2$. Chemical instability occurs when three real roots exist, two of which will be positive, one negative. One negative root is present because c_0/c_3 is positive. When Coulomb effects are turned off $(C=0)$, at $y=1/2$, the two positive solutions are the same as that for mechanical instability. When y is no longer 1/2, the mechanical instability and chemical instability regions are no longer the same. This result is shown in the lower part of Figs. 1 and 2 for various choices of y. It should also be noted, for $C=0$, that the coefficients of the cubic equation have a certain symmetry arising from the form in y which is $y(1-y)$, i.e., interchanging y (protons) and $1-y$ (neutrons) leaves the solution invariant [see curves for $(d\mu_q/d\rho)_{y,T}=0$ in Figs. 1 and 2]. This result is no longer true when Coulomb interactions are included.

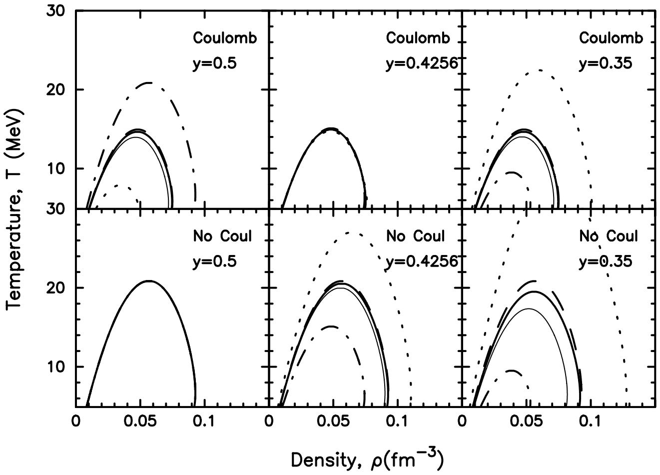

FIG. 2. The temperature dependence of the chemical and mechanical instability curves at a fixed proton concentration y. The curves here are the constant y plane cuts of the corresponding surfaces in (ρ,y,T) space. The curves are the same as in Fig. 1 except the dashed curves. With the temperature dependent equal concentration $y_E(T)$, all the curves coincide as shown by a dashed line here [note here that $y_E(T)=1/2$ without Coulomb force].

We now turn our attention to the simple equation of state and μ_q, but now with asymmetric Coulomb effects included $(C\neq0)$ in a simplified form which will illustrate its qualitative effects. The calculations shown in Figs. 1 and 2 use Eqs. (53) and (54) of Ref. [4] for μ_q and P with degeneracy correction representing a first-order quantal effect. The mean field force parameters are the same as used in Ref. [4] except for using $C_S=0$ (surface term) and using a larger radius $R=8$ fm for a larger C (Coulomb term). C, which depends on R, is taken to be a constant. For this choice, we can consider R as an effective range of the Coulomb force, which is now independent of the density. This leads to a Coulomb interaction that depends on the square of the proton density. This simplification reduces the complexity of many results. To make the Coulomb effects more visible in our figures (Figs. 1 and 2), we use a large value of $C=231$ MeV/fm^3 which is appropriate for systems near U. Smaller systems will have a smaller C. Also, the valley of β stability is closer to the equal concentration line for smaller systems.

Some qualitative features of the effects of such a Coulomb behavior are the following (see Figs. 1 and 2 also).

(1) The mechanical (thin solid line) and chemical (thick solid line) instability curves are the same when $(dP/dy)_{\rho,T}=0$ which occurs at $y_E=0.5/(1+C/[(\frac{8}{3})(\frac{1}{2}+x_0)a_0])$ and thus no longer at $y_E=0.5$ when $C\neq0$. This equation for the proton concentration of y_E, which is on the line of equal concentration on the coexistence surface [4], has a similar structure to the equation that gives the Z which has the maximum binding energy at a fixed A (valley of β stability) obtained from the liquid drop mass formulas or from Eq. (55) of Ref. [4]. With higher-order T dependence coming from degeneracy corrections, y_E depends on T and $y_E=0.4256$ for $x_0=\frac{1}{2}$ at $T=10$ MeV. Explicitly, with first-order degeneracy corrections, $y_E=0.5/(1+C/[(\frac{8}{3})(\frac{1}{2}+x_0)a_0$

$+T\lambda^3/(2\sqrt{2}\gamma)]$). The curves of $(d\mu_q/d\rho)_{y,T}=0$ also coincide with the instability boundary at y_E. While all the curves coincide at $y=\frac{1}{2}$ (without Coulomb) as shown in the lower left box of Fig. 2, the top middle box of Fig. 2 with Coulomb (even though it is hard to distinguish) exhibits a separation between these curves for a fixed $y=0.4256$ due to the temperature dependence of $y_E(T)$. All the four surfaces in (ρ,y,T) space coincide at the line of temperature dependent $y_E(T)$ and that the mechanical instability surface is inside of the chemical instability surface.

(2) For both larger and smaller values of y from y_E, the mechanical instability boundary (thin solid line) is inside of the chemical instability region (thick solid line) and these coincide at $y_E(T)$ (Fig. 1 and upper middle box of Fig. 2). Upper part of Fig. 2 and upper left box of Fig. 1 also show that the roles of $(d\mu_p/d\rho)_{y,T}=0$ and $(d\mu_n/d\rho)_{y,T}=0$ are exchanged when y crosses y_E. The mechanical instability has its peak in T versus ρ at $T_c^P(y)=[a_0-(2/3)(1/2+x_0)a_0(2y-1)^2-Cy^2]^2/(6a_3)$, which is maximum at $y=y_E$ and coincide with chemical instability at y_E.

(3) The maximum critical T_c obtained from the peak in the chemical equilibrium surface of T versus ρ occurs at $y\neq 0.5$, but is now shifted to $y=y_E(T)$. The coexistence boundary coincides with the chemical instability boundary at the line of critical points $y_c(T)$ (peaks in Fig. 1 for a $T=10$ MeV plane cut), but not with the mechanical instability boundary.

(4) At $y=y_E(T)$, the equilibrium curve or binodal curve of P versus y obtained by cutting the binodal surface at a given T meets at a point, i.e., the liquid and gas phases have the same proton concentration (see Fig. 4 of Ref. [4]). When Coulomb effects are turned off, the binodal section meets at $y=1/2$. At $y=y_E(T)$ with Coulomb effects included a two-component system will behave as a one-component system, keeping the proton/neutron ratio the same in both liquid and gas phases. These results indicate that a slowly expanding system reaches the diffusive chemical instability region before reaching the mechanical instability region except for the case of proton concentration y_E. If the system has a proton concentration smaller than y_E, then neutron diffusion occurs, while proton diffusion occurs for a system with $y>y_E$. If the system has a proton fraction of y_E, then the phase transition is purely mechanical without chemical diffusion. Since the equal concentration y_E is similar to the proton concentration for the valley of β stability, β stable nuclei would experience mostly a mechanical phase transition rather than a chemical phase transition. Radioactive beam with more protons than in β stable nuclei diffuses protons dominantly making the proton fraction smaller in the liquid phase, while nuclei with more neutrons than in β stable nuclei diffuse neutrons dominantly leaving a bigger proton fraction in the liquid.

In summary, we have presented calculations based on a mean field theory with asymmetry, which include the role of Coulomb force on the liquid-gas phase instability regions for both the mechanical (spinodal) surface and the chemical (diffusive) surface. The asymmetric Coulomb force modifies these surfaces so that their peaks are no longer at proton fraction $y=1/2$ but shifted to a smaller value called y_E which is close to the proton fraction in the valley of β stability. The robustness of the isospin fractionization is somewhat moderated by the presence of the Coulomb force. The mechanical instability surface is always inside the chemical instability surface, meaning that the diffusive chemical transition occurs even in a mechanically stable region. The two surfaces are the same at the equal concentration y_E (allowing for a quantal T dependence in y_E). At y_E, the peaks in the chemical instability surface and binodal equilibrium surface also coincide. For systems with $y\neq y_E$, the peaks in the T versus ρ curve for chemical instability and chemical equilibrium—binodal or coexistence curve—coincide, but the peak for mechanical or spinodal instability is below that of the other two. At $y=y_E$, a phase transition in a two-component system behaves the same as a one-component system, keeping the proton fraction at y_E in both phases during the entire transformation. For a system with $y<y_E$, neutron diffusion is important, while proton diffusion dominates for a system with $y>y_E$. Rare isotope accelerators have the possibility of studying some of these features. Finally, finite nuclei offer the possibility of studying phase transitions in two-component systems where volume, surface, symmetry energy, asymmetric or Coulomb energy, and quantal effects play an important role. Such studies give additional information about the general behavior of these transitions. We have tried to show these features in a simplified analytic model, where isospin symmetries and isospin invariance ($y\leftrightarrow 1-y$) are broken in a transparent way with an asymmetric Coulomb interaction.

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