Liquid-gas phase transition and its order in finite nuclei

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Abstract

The liquid-gas phase transition in finite nuclei is studied in a heated liquid-drop model where the drop is assumed to be in thermodynamic equilibrium with the vapour emanated from it. Changing pressure along the liquid-gas coexistence line of the systems, symmetric or asymmetric, suggests that the phase transition is a continuous one. This is further corroborated from the study of the thermal evolution of the entropy at constant pressure.

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The study of liquid-gas phase transition in finite nuclear systems is of considerable contemporary interest [1,2,3,4,5,6,7,8,9]. Experimental analyses of the accumulated data on multifragmentation and caloric curves show compelling evidence of such a transition. Phase transitions are normally signalled by peaks in the specific heat at constant volume $C_V$ with rise in temperature. Theoretical models of different genres, such as the microcanonical [10] or the canonical [11] description of multifragmentation, the lattice-gas model [7,8,12] or even the microscopic treatment in a relativistic [13] or a nonrelativistic [3] Thomas-Fermi framework support such a structure in the heat capacity. A clear idea about the subtle details of the liquid-gas phase transition in finite nuclei, however, has not emerged yet. Confusion remains about whether the system has evolved dynamically through the critical point [6,14,15]; a coherent picture about the order of the phase transition is also missing. Analyses of the EOS group [16] and the ISiS group [17] in the scaling model give strong circumstantial evidence for a continuous (second order) phase transition. Calculations performed in a mean field model [5] also lead to a similar conclusion. Predictions from the lattice-gas models [7,18] are, however, compatible with a first order transition.

Symmetric infinite nuclear matter is effectively a one-component system; with heating, it undergoes a first order phase transition. On the other hand, in case of two-component asymmetric nuclear matter, as was shown in a comprehensive analysis by Müller and Serot [19] in a relativistic mean-field framework, the separate conservation of the neutron and proton number densities leads the system to a continuous phase transition over a finite temperature interval. This is also supported in the nonrelativistic calculations by Kolomietz et al [20]. Unlike symmetric nuclear matter, even a finite symmetric nucleus ($N = Z$) behaves like a two-component system as the Coulomb interaction lifts the isospin degeneracy. A finite nucleus is then expected to undergo a continuous liquid-gas phase transition, if at all. The
conflicting predictions from the previous model analyses do not have realistic inputs of nuclear physics as relevant for a quantum system of interacting fermions with a short-range nuclear force as also with the long-range Coulomb interaction. In this communication, we focus on the nature of the phase transition once specific features concerning an atomic nucleus are properly taken into account.

For our study, we choose a representative system, namely, Rhenium with $A = 186$ and $Z = 75$. Such a nucleus is likely to be formed from the reaction $^{124}Sn + ^{124}Sn$ at energies around 50 MeV per nucleon [21] after some nucleons have left out from the reaction zone as preequilibrium particles. We also investigate $^{150}Re$ to explore the isospin asymmetry effects in phase transition. The nucleus is viewed as a spherical liquid drop with asymmetry $X_0$ defined as $(N_0 - Z_0)/A_0$ where $A_0 = N_0 + Z_0$, is the mass number of the total system. At a finite temperature, we assume the depleted nucleus to be enveloped by its own vapour and the system to be in complete thermodynamic equilibrium conserving the total number of neutrons and protons. The nucleon distributions in the liquid and gas are assumed to be uniform in each phase. This definition allows to explore the liquid-gas coexistence region for a finite system in close analogy with bulk nuclear matter.

In absence of a well-defined way to write the energy density functional of a finite nucleus in terms of volume, surface, symmetry and Coulomb terms, we write the free energy of the nuclear system at temperature $T$ in the single phase as

$$F = A_0 f_{nm}(\rho, X_0) + F_C + F_{surf}$$  \hspace{1cm} (1)

where $f_{nm}(\rho, X_0)$ is the free energy per particle of infinite nuclear matter at density $\rho$ with asymmetry $X_0$ at the same temperature $T$, $F_C$ the Coulomb free energy and $F_{surf}$ the temperature and asymmetry dependent surface free energy. In the liquid-gas coexistence region, the free energy is given by

$$F_{co} = F^l + F^g,$$  \hspace{1cm} (2)
where the liquid free energy is,

$$F^l = A^l f_{nm}(\rho^l, X^l) + F^l_C + F^l_{surf},$$

and the free energy of the emanated gas is

$$F^g = A^g f_{nm}(\rho^g, X^g) + F^g_C.$$  (4)

Here $A^l$ and $A^g$ are the number of nucleons in the liquid and the gas phase, $\rho^l, \rho^g$ and $X^l, X^g$ are the corresponding density and asymmetry. The free energy of infinite nuclear matter is evaluated in the finite temperature Thomas-Fermi framework with a modified Seyler-Blanchard interaction [22]. The Coulomb free energies for the liquid and the gas, $F^l_C$ and $F^g_C$ are calculated corresponding to a uniform charged sphere and a spherical-shell, respectively. For simplicity, their mutual interaction is neglected. The surface free energy of the liquid part $F^l_{surf}$ is taken as [23]

$$F^l_{surf} = \sigma(X^l, T) (A^l)^{2/3},$$

where the surface energy coefficient is

$$\sigma(X, T) = [\sigma(X = 0) - a_s X^2][1 + 1.5T/T_c][1 - T/T_c]^{3/2}. \quad (6)$$

Here, $\sigma(X = 0) = 18$ MeV, $a_s = 28.66$ MeV and $T_c$, the critical temperature of the symmetric nuclear matter is 15 MeV. The surface energy coefficient decreases with density; we neglect its density dependence for the liquid part. Since the gas density is very low, its surface energy is neglected. The total surface energy of the liquid is

$$E^l_{surf} = F^l_{surf} + T S^l_{surf},$$

where the total surface entropy is given by

$$S^l_{surf} = - \left( \frac{\partial F^l_{surf}}{\partial T} \right)_{V}. \quad (8)$$
The total entropy $S_0$ of the system is then calculated as

$$S_0 = A_l s_{nm}^l(\rho_l, X_l) + A_g s_{nm}^g(\rho_g, X_g) + S_{surf}^l.$$  (9)

The per particle entropy $s_{nm}$ of homogeneous nuclear matter is obtained from the standard mean-field prescription.

The chemical potentials $\mu_n^l$ and $\mu_z^l$ for neutron and proton in the liquid are given by

$$\mu_n^l = \frac{\partial F^l}{\partial N_l^l},$$
$$\mu_z^l = \frac{\partial F^l}{\partial Z_l^l}.$$  (10)

The liquid pressure is obtained from

$$P = \rho^2 \frac{\partial (F/A)}{\partial \rho}.  \quad  (11)$$

Similar equations follow for the gas phase. For thermodynamic equilibrium between the liquid and the gas, the two chemical potentials and the pressures in both phases must be the same, i.e., $\mu_n^l = \mu_n^g$, $\mu_z^l = \mu_z^g$ and $P^l = P^g$. Along the coexistence line, the mass of the liquid drop changes. For a chosen $A_l^l$, the quantities $\rho_l^l$, $\rho_g^g$, $X_l^l$, $X_g^g$ and $A_g^g$ are determined by exploiting the three thermodynamic equilibrium conditions and the constraints of baryon number and the total isospin conservation:

$$\rho_{n,z} = \lambda \rho_{n,z}^l + (1 - \lambda) \rho_{n,z}^g,$$
$$\rho X_0 = \lambda \rho^l X^l + (1 - \lambda) \rho^g X^g.$$  (12)

Here $\rho$ is the average nucleon density and $\lambda$ is the liquid volume fraction.

The isotherms for the nucleus $^{186}\text{Re}$ at $T = 7, 8, 9$ and 10 MeV are displayed in Fig.1. For comparison, the isotherm for nuclear matter with asymmetry same as that of $^{186}\text{Re}$ is also shown at $T = 10$ MeV. The difference between the isotherms for the infinite and the finite system is not insignificant. Though the Coulomb and
surface have opposing effects on the pressure, the former wins over the latter at this temperature. The liquid-gas coexistence lines for $^{186}$Re for the four temperatures mentioned are shown by the dotted lines. It is seen that the pressure changes along the coexistence line, as seen earlier in the case of asymmetric nuclear matter [19]; the slope of the coexistence lines also increases with temperature. It is further noted that at a given temperature and asymmetry, because of the Coulomb effect, the slope of the coexistence line of a finite nucleus is more compared to that of asymmetric nuclear matter. The variation of pressure along the coexistence line is a pointer to a continuous phase transition. Unlike nuclear matter, the coexistence lines do not extend from the pure gas phase to the pure liquid phase. At relatively lower temperatures, it is found that as the system expands, the size of the liquid drop depletes and reaches a minimum mass beyond which no thermodynamic equilibrium is possible; at higher temperatures, with compression the liquid drop attains a limiting mass which decreases with increasing temperature as is evident from the figure. As an example, the minimum liquid-drop mass at $T = 7$ MeV is $A_l = 24$ (marked as A in the figure); at $T = 10$ MeV, the limiting liquid-drop mass is $A_l = 130$ (marked as B). This implies that at lower temperatures, a gas of a finite number of nucleons when compressed start nucleating with a minimum mass for the seed in order to remain in thermodynamic equilibrium. Similarly, at higher temperatures, for the coexisting finite system, the evaporated gas should contain a minimum number of nucleons.

The isospin fractionation along the coexistence line for the nucleus $^{186}$Re at $T = 8$ MeV is shown in Fig.2. The system has proton fraction $Y_0 = 0.403$ (defined as $Y = Z/A$). As the system prepared in the gaseous phase is compressed, the two-phase region is encountered at the point A with the emergence of a minimum liquid mass ($A_l = 20$) at the point B with a proton fraction $Y_B$ larger than $Y_0$. With further compression, the gas phase depletes from A to C while the liquid phase grows from
B to D attaining the total mass $A_0$ and proton fraction $Y_0$. During compression, the proton fractions in both phases decrease, but the total proton fraction $Y_0$ remains fixed, as dictated by the conservation of the total isospin given by Eq.(12). It is evident from the figure that the gas phase is more neutron-rich compared to the total system while the liquid phase is comparatively neutron-deficient as also observed experimentally [21]. This feature becomes more prominent with increasing liquid mass. In order to explore the asymmetry effect on isospin fractionation in phase transition, the calculated results for the symmetric system $^{150}$Re are also displayed in the figure. Contrary to the relatively neutron-rich nucleus $^{186}$Re, here it is found that the gas phase is proton-rich. In symmetric nuclei, since $\mu_z$ is greater than $\mu_n$ because of Coulomb interaction, the separation of the gas phase behaves more like that of proton-rich nuclear matter. The occurrence of liquid-gas phase transition in symmetric medium-heavy nuclei should then lead to preponderance of proton-rich isotopes in energetic heavy-ion collisions and this can be tested in experiments.

The heat capacity per particle $C_V$ for $^{186}$Re at constant volume (defined as $\left(\frac{d(E^*/A_0)}{dT}\right)_V$, where $E^*$ is the total excitation energy of the system) is displayed in Fig.3 at a representative volume $V = 10V_0$ which can be interpreted as a freeze-out volume. Here $V_0$ is the normal volume of the nucleus calculated with the radius parameter $r_0 = 1.16$ fm. A very broad bump in $C_V$ with a maximum at $T \sim 10$ MeV is seen. The system then corresponds to a liquid part with $A_l$ around 80, the rest of the nucleons being in the gas phase. This is contrary to the results in the microscopic mean-field calculations obtained earlier [3,13] at around the same freeze-out volume, where a much sharper peak was observed when the system just vaporises completely. At very high temperature, the heat capacity saturates at 1.5 corresponding to a pure classical gas.

The thermal evolution of the entropy per particle for the nucleus $^{186}$Re at a constant pressure $P = 0.06$ MeV fm$^{-3}$ is shown by the solid line in figure 4. In contrast
to a first order phase transition where the entropy at constant pressure exhibits a discontinuity at a particular temperature (the phase transition temperature), here the entropy change is continuous. The noticeable rise in entropy in the temperature range 8.4 to 10.3 MeV is the manifestation of a liquid-gas phase transition in this temperature domain for the chosen pressure. The dashed line in the figure corresponds to the entropy evolution for the symmetric nucleus $^{150}$Re. This is quite similar to that of the asymmetric nucleus considered, but markedly different from that of symmetric nuclear matter $^{19}$ which shows a sharp discontinuity at the transition temperature. The striking similarity between the two results shows that the finite size and Coulomb effects are the dominant factors in determining the nature of phase transition in finite nuclei. The continuous change of entropy at a constant pressure plead in favour of the characterisation of the liquid-gas phase transition in atomic nuclei, symmetric or asymmetric, as a continuous one.

The liquid-gas phase transition in finite nuclei with explicit conservation of the baryon number and the total isospin has been investigated in this communication in a heated liquid drop model. The peaked structure in the heat capacity, though broad, signals the occurrence of a liquid-gas phase transition. From the evolution of entropy at constant pressure, one sees that the transition occurs over a range of temperatures; this strongly suggests that the liquid-gas phase transition in a finite nuclear system is continuous. The simplified assumptions in the model may affect the results somewhat quantitatively, but the general qualitative features are expected to remain unaltered. The thermodynamic concepts may not be very meaningful when the number of particles in one of the phases is very small, still this model serves as a window to understand the basic features of liquid-gas phase transition in finite systems.
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Figure Captions

Fig. 1 The isotherms for the system $^{186}Re$ at different temperatures as labelled on each curve are shown as full lines. The liquid-gas coexistence lines are shown by the dotted lines. The long-dashed line refers to the isotherm at $T = 10$ MeV for nuclear matter with asymmetry same as that of $^{186}Re$. The corresponding coexistence line is shown by the filled circles.

Fig. 2 Evolution of proton fraction ($Y$) along the liquid-gas coexistence line at $T = 8$ MeV for the systems as shown.

Fig. 3 The specific heat capacity $C_V$ as a function of temperature for $^{186}Re$ in a freeze-out volume $10V_0$.

Fig. 4 Entropy per particle as a function of temperature at a constant pressure for the systems $^{186}Re$ and $^{150}Re$. 
$P \ (\text{MeV/fm}^3)$

$\rho \ (\text{fm}^3)$

Fig. 1
Fig. 2
Fig. 3
**Fig. 4**

$P = 0.06 \text{ MeV fm}^{-3}$

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- Dashed line: $^{150}\text{Re}$
- Solid line: $^{186}\text{Re}$