Nuclear (multi-)fragmentation

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Abstract. Nuclear (multi-)fragmentation, defined as the nuclear decay mechanism in which at least three intermediate mass fragments \((Z \geq 3)\) are produced, is the disassembly phenomenon specific to hot nuclear matter produced in nuclear collisions at beam energies of 20-100 MeV/nucleon. Considered as the manifestation of the liquid-gas phase transition predicted by nuclear matter mean-field models or an instrument to address the nuclear equation of state, it enjoys for thirty years a vivid scientific interest and motivates the construction of more and more high-performance \(4\pi\)-detectors allowing for almost complete reaction characterisation on an event-by-event basis. Over the years, many experimental signatures of first- and second-order phase transitions have been proposed and identified in both central and peripheral collisions. Despite their seemingly puzzling messages, coherent understanding of multifragmentation is possible accounting for statistical ensemble inequivalence and finite size effects. Two particular situations will be discussed in detail. The first one corresponds to a multifragmenting medium size nucleus where the expected first-order phase transition is blurred by surface effects. The second one corresponds to an infinitely large system, the clusterized nuclear matter thought to constitute the main sector of baryonic matter in (proto-)neutron stars, where the first-order phase transition is quenched by Coulomb effects. In both cases statistical models with cluster degrees of freedom have been employed.

1. Introduction

Due to the short-range repulsive and finite-range attractive features of the nucleon-nucleon interaction, the idea that nuclear matter exhibits a phase diagram similar to the liquid-gas one was advanced already thirty years ago \([1, 2]\). Detailed studies performed within mean-field approaches have confirmed the presence of first and second-order phase transitions and determined the evolution of the phase coexistence domain with respect to temperature and isospin asymmetry \([3, 4, 5, 6]\). Moreover, the existence of a density-driven instability domain where density fluctuations are spontaneously and exponentially amplified in order to achieve phase separation lead to the idea that over the corresponding sub-saturation density range the homogeneous matter scenario cease to stand valid and matter organizes itself in clusters.

Clusterized structures of dilute nuclear matter potentially stemming from the spinodal instabilities were experimentally evidenced in nuclear multifragmentation, that is the decay mechanism taking place at excitation energies of the order of 2-10 MeV/nucleon \([7]\), and an impressive experimental and theoretical effort was done with the aim to answer the challenging question on whether this decay channel really corresponds to a phase transition or not. The difficulties of this task were both conceptual and technical. From the conceptual point of view, the finite size of a nucleus makes it lie outside the domain of validity of the phase
transition theory which is the thermodynamical limit (i.e. infinite size). This means that in the most optimistic situation in which a nucleus would behave like the infinite, electrically uncharged, homogeneous system that nuclear matter is, none of the known first or second order phase transition signals could survive undistorted in multifragmentation. The situation is even more dramatic if one takes into account that, for a charged system, the thermodynamical limit is, by construction, not achievable because of the diverging Coulomb energy. Moreover, Coulomb and surface interaction energies which scale with $Z^2$ and, respectively, $A^{\sigma}$, violate extensivity property and might be responsible for statistical ensemble in-equivalence, that is the dependence of the observed physics on the constraints externally imposed to the system. From the experimental perspective, the situation is equally difficult given that the experimentally accessible information concerning fragment partitions and kinetic properties does not correspond to the thermodynamically relevant break-up stage, but to the so called asymptotic one obtained after the break-up: hot fragments suffered a sequence of particle evaporation processes and propagation in the mutual Coulomb field. Not less important, most intensive thermodynamical observables (temperature, pressure, density) which are essential in plotting the phase diagram or characterize the path followed by the system in the phase space are subject to large uncertainties or impossible to constrain.

All these issues have been thoroughly addressed over the last two decades and led, on one hand, to the re-formulation of phase transition theory in the case of finite systems [8] and, on the other hand, to the construction of performant $4\pi$-detectors allowing for almost complete reaction characterization on an event-by-event basis [7, 9].

What is interesting is that the efforts paid in order to understand the thermodynamics associated to multifragmentation pay their dividends in helping us to interpret the thermal and phase properties of dilute unhomogeneous baryonic matter produced in the explosion of core-collapsing supernova and neutron star crust. At variance with a fragmenting nucleus, star matter is - because of the electrons - neutral and, practically, at the thermodynamical limit. Though, phase separation is quenched as the huge electron gas incompressibility prevents the electrons to follow on a macroscopic scale the baryonic matter density fluctuations such as to compensate the proton charge. As such, dishomogeneities are of finite size and the whole can be viewed as an admixture of nuclear clusters and nucleons [10]. Mastery of both the thermodynamical response of baryonic matter, commonly termed as equation of state (EOS), and its chemical composition is of major astrophysical interest as it influences the dynamics of core-collapsing supernovae and cooling phase of proto-neutron stars.

In the following we shall spot the thermodynamics of multifragmenting nuclei and dilute star matter within a statistical model with cluster degrees of freedom, commonly known under the generic name of nuclear statistical equilibrium (NSE) models. This kind of approaches rely on the hypotheses that nuclear correlations are entirely exhausted by clusterization and offer the remarkable advantage of treating all nuclear bound and continuum states via empirical parameterizations. Their major inconvenience is the neglect of in-medium effects. In order to approach the crust-core transition and account for nucleon-nucleon interactions in the vicinity of normal nuclear density and beyond that, in the case of star matter the loosely interacting gas of clusters is considered to coexist with an uniform gas of interacting nucleons treated within the mean-field approach.

We shall first show that an infinite neutral clusterized system manifests first and second order phase transitions. Then, we shall demonstrate that, because of surface, for a multifragmenting nucleus in which Coulomb interaction is switched off the first order phase transition is quenched but it can be traced back from the back-bending behavior of any $Y_i(X_i)|_{X_{j},j\neq i}$ curve or, equivalently, bi-modal pattern of probability distributions of the order parameters of the phase transition. $Y_i$ and $X_i$ stand for an intensive and, respectively, the conjugated extensive variable. Further on we shall show that provided that Coulomb effect is not too strong, the fossil signal
of the phase transition can be found also in real (charged) nuclei. Finally, we shall show that the non-saturating Coulomb interaction is responsible for statistical ensemble in-equivalence in non-homogeneous dilute star matter and that correct thermodynamical characterization can be achieved only in the ensemble in which the order parameter of the phase transition, i.e. total baryonic density, is controlled, i.e. the canonical ensemble.

2. Infinite neutral clusterized matter
Let us first consider a one-component clusterized gas of non-interacting clusters whose energy density functional $B(A) = a_0 A - a_s A^{2/3}$ consists, similarly to the one of nuclei, of a bulk and a surface term and deduce, under the equilibrium hypotheses, the associated thermodynamics.

Probability distributions of the total number of particles and energy, the expected order parameters of a liquid-gas (LG) type phase transition, of a system composed of an arbitrary number of $A_0=2000$ particles occupying an arbitrary volume $V=14476.4$ fm$^3$ at $T = 8.3, 10.35, 13.19$ MeV as obtained out of a grand-canonical Monte-Carlo Metropolis simulation are plotted in Fig. 1 (left and right panels). As one may note, both variables present a bi-modal pattern meaning that the system manifests a first-order phase transition. It comes out that the dense phase is characterized, whatever the temperature $T < T_C$, by a density equal to the normal nuclear matter density. At contrast, the dilute phase frontier evolves monotonically with the temperature such that the phase coexistence region shrinks (open symbols). At a certain value of the temperature, $T_C \approx 16$ MeV, it gets reduced to a unique point, the second-order phase transition. As such one may conclude that, as expected given the van der Waals-type of the interaction, clusterized nuclear matter behaves like macroscopic fluids. A difference is nevertheless worthwhile to be noticed: the limiting point of the phase diagram is characterized by $\rho = \rho_0$ which suggests that it does not correspond to the critical point of the Fisher model [11]. This fact is confirmed by the study of critical exponents, as demonstrated in Ref. [12]. What is more remarkable and encouraging for experimentalists aiming to pin down the phase diagram out of multifragmentation data is that the largest fragment bi-modal distribution correctly signals phase coexistence (middle panel in Fig. 1): the different temperatures for which the two peaks

![Figure 1](image-url)
corresponding to the two phases have the same height differ by less than 0.1 MeV from the ones corresponding to equal height peaks in $P(E)$ and $P(\rho)$. This is obviously due to the correlation between $A_{\text{max}}$ and the system’s total energy.

3. Un-charged multifragmenting nucleus

To what extent the large system phase transition phenomenology stands valid while decreasing the system size is a non-trivial question taking into account that surface effects and conservation laws play a non-negligible role. Surface effects, which do not scale with the system size, are expected to violate extensivity. Finite size and conservation laws are expected to induce statistical ensemble in-equivalence meaning that, in order to pin down the phase coexistence, one has to choose a signal compatible with the way in which the system is constrained. To account as realistically as possible for the particularities of an isolated system exploding into vacuum, it is clear that the most appropriate statistical ensemble is the microcanonical one. The details on how microcanonical multifragmentation is modeled and numerically solved may be found in Ref. [13]. But, constraining the energy, volume and particle number, that is the order parameters, does not allow the system to explore the two phases provided that the external constraints are such that it lies in the phase coexistence region. To overcome this inconvenient, one has to work in a constant-pressure canonical ensemble. One possibility to do that is to start from microcanonical configurations and alter accordingly their statistical weights,

$$P(E, V) = \frac{W(E, V) \exp(-\beta E - \beta PV)}{Z_{\text{can}}(\beta, P)},$$

where $W(E, V)$ stands for the microcanonical weight of a micro-state defined by the excitation energy $E$ and freeze-out volume $V$, $P$ is the externally imposed pressure, $\beta = 1/T$ is the inverse temperature and $Z_{\text{can}}(\beta, P)$ is the canonical partition function.

The right-upper panel of Fig. 2 depicts (in log scale) the probability distribution $P(E, V)$ corresponding to the un-charged nucleus (200, 82) at $T = 6.678$ MeV and $P = 2 \times 10^{-2}$ MeV/fm$^3$. 

Figure 2. Right-upper panel: probability distribution (in log scale) of a canonical ensemble at constant pressure corresponding to the un-charged nucleus (200, 82). The thick lines correspond to microcanonical isobaric and isothermal paths corresponding to $P_\mu = P$ and $T_\mu = 1/\beta$. The lateral panels correspond to projections (dashed lines) of the probability distributions on $V/V_0$ (left) and excitation energy $E$ (down) axes. Canonical pressure (left panel) and constant-$\lambda$ temperature are figured with thin lines. The corresponding microcanonical curves are plotted with thick lines. Figure taken from Ref. [14].
The obvious bi-modal pattern as a function of both excitation energy and volume stems from the convexity anomaly of the associated thermodynamical potential $\bar{S}[T,P]$, where $\bar{S}[Y_i]$ represents the Legendre transformation of the entropy with respect to the fixed intensive $Y_i$, and signals phase coexistence. In addition to how this two-dimensional probability distribution looks like while projected on the two axes $E$ and $V$ (dotted lines), the lateral panels show a constant-pressure caloric curve $T(E)|_P$ (lower panel) and, respectively, a constant-temperature pressure versus volume curve $P(V)|_T$ (left panel). For the sake of completeness, also constant $\lambda = \beta P$ caloric curves and pressure versus volume curves are represented. All these curves back-bend in the region situated between the peaks of the probability distribution which corresponds, in the mean-field approach, to the spinodal instability domain.

In conclusion, 1) a neutral finite system keeps the memory of the LG phase transition taking place in infinite matter, 2) first-order phase transition is quenched by surface effects and 3) back-bending behavior of $T(E)|_P$ and $P(V)|_T$ is a signature of phase coexistence.

4. Multifragmenting nucleus

Despite the achieved progress in understanding in what regards a finite system differs from its infinite counterpart, our main question is still not answered because, as already stressed, real nuclei are also electrically charged. At first glance this should imply, in the most optimistic scenario, a decrease of the limiting temperature and, in a more pessimistic one, the complete suppression of phase coexistence.

Using the previously discussed back-bending behavior of isobar caloric curves, Fig. 3 shows that at least for relatively small nuclei, where Coulomb field is not too strong, the phase transition survives.

5. Dilute star matter

Clusterized nuclear matter is not only created in small amounts in laboratories employing heavy ion collisions at bombarding energies of the order of several tens of MeV/nucleon but is abundantly populated during the post-bounce phase of a core-collapsing supernovae, as well as over the much longer Kelvin-Helmholtz epoch during which the remnant changes from a hot lepton-rich proto-neutron star to a cold deleptonized neutron star. The duration of the first process is of about several hundreds of ms while the one of the latter is of several tens of seconds. In both cases temperatures $10^9 < T < 2 \cdot 10^{11}$ K, nuclear sub-saturation densities
Figure 4. Symbols: Grand-canonical (left) and canonical (right) $\mu_{\text{tot}}(\rho_{\text{tot}})$ at $T = 1.6$ MeV and $\mu_I = 1.68$ MeV. Vertical dashed line illustrates the Gibbs construction; horizontal dot-dashed lines mark the frontiers of the phase coexistence domain. Figure taken from Ref. [17].

$10^{10} < \rho < 10^{14}$ g/cm$^3$ and electron fractions $0 < Y_e < 1$ are spanned.

Adding to the previously discussed loosely interacting gas of clusters an extra component made out of an uniform distribution of interacting nucleons such as to correctly describe the crust-core solid-liquid transition phenomenology, we investigate whether phase coexistence really occurs in star matter [16, 17]. The fragment component is described by a liquid drop energy density functional whose parameters are fitted to experimental data but no restriction is imposed with regard to the cluster size or isospin-asymmetry and the Coulomb term is altered such as to account for the screening effect of the electrons. The latter is estimated within the Wigner-Seitz approximation. For the uniform gas of nucleons we use a mean-field approach and Skyrme-like interaction potentials. The assumed lack of interaction between the two components otherwise than via the excluded volume allows the total partition function be factorized in a free nucleon and, respectively, a cluster gas component such that the total entropy writes

$$
\sigma_{can,\beta,\mu_I}^\text{can} (\rho) = \ln Z_{\beta,\mu_I}^1 (\rho_f) + \lim_{V \to \infty} \frac{1}{V} \ln Z_{\beta,\mu_I}^{\rho_d > 1} (V \rho_{\text{cl}}).
$$

(2)

Taking into account that 1) in a two-component system a standard procedure of reducing the mathematically difficult Gibbs construction to a much easier Maxwell construction is to control one of the two chemical potentials and the other particle density and 2) at constant temperature the phase separation direction in NM is largely dominated by the total density, we choose to fix the iso-vector chemical potential $\mu_I = (\mu_n - \mu_p)/2$.

The evolution of total baryonic density with respect to the conjugated total chemical potential $\mu = (\mu_n + \mu_p)/2$ as obtained in a grandcanonical ensemble is represented with open symbols in the left panel of Fig. 4 for $T=1.6$ MeV and $\mu_I = 1.68$ MeV. As one may note, there is an important density range which is jumped over and, for a certain domain of $\mu$, two kinds of mixtures exist. One of them entirely consists of clusters. The second one consists of uniform NM at normal or supra-saturation nuclear density. The impossibility to access the phase coexistence density range while controlling the intensive conjugated to the order parameter is a notorious issue in the case of first-order transitions. Which of the multiple mixture solutions obtained for $-16 < \mu < -9.5$ MeV or a linear combination of them corresponds to equilibrium can be learnt by seeking in which case the associated energetic thermodynamical potential gets minimized. The result is plotted with a vertical dashed line on the figure and corresponds to the Maxwell construction performed for $\mu \approx 16$ MeV. This means that the dilute clusterized phase obtained for larger values is meta-stable.
Taking into account that 1) charged matter can occur on macroscopic scales if and only if net charge neutrality is strictly obeyed, 2) because the symmetry energy, a density fluctuation leads to a proton, \textit{i.e.} charge, fluctuation, 3) electron gas incompressibility might not allow them to follow the baryonic matter fluctuations such as to compensate the proton charge, one may wonder whether phase separation may really exist.

The answer can be given by following how the system behaves while constraining the density, that is in a canonical ensemble. If the thermodynamics was that of a first-order phase transition, a plateau would be obtained. If, by contrary, the system preferred to organize itself in an admixture of free nucleons and nuclear clusters, the transition between the extremities of the Maxwell construction would be continuous. The right panel of Fig. 4 confirms the last scenario meaning that the dis-homogeneities happen at microscopic scale. From the statistical physics point of view, the situation is a paradigm of ensemble in-equivalence and represents, to our knowledge, one of the few cases in which it could have significant phenomenological consequences. The narrow density interval presenting the most sudden evolution of $\mu(\rho)$ and whose extremities are marked by stars corresponds to the so-called pasta phase region disregarded by the spherical symmetry assumed in our simple model.

6. Conclusions
Clusterized structures in fragmented excited nuclei and dilute star matter are discussed from the perspective of statistical models with cluster degrees of freedom. It was shown that multifragmentation does really correspond to a phase transition but phase separation is quenched by finite size effects. In the case of dilute stellar matter, phase separation is also quenched and the first-order phase transition experienced by nuclear matter is replaced by a continuous one.

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