4- Instabilities and fragmentation

1. The dynamics of cluster formation

In the previous lesson (course 3) we have shown that, if we consider nuclear matter inside the spinodal region, the fact of imposing a density fluctuation to an initially homogeneous system leads to a reduction of the free energy density. We have interpreted this behavior as the signature that the system is unstable with respect to density fluctuations, and that this instability will give rise to cluster formation and/or phase separation. In this chapter we develop this statement by studying the time evolution of density fluctuations.
Let us consider for simplicity again the case of a single density variable. This can still represent a nucleus composed of neutrons and protons, if we assume that the isospin degree of freedom is frozen and the fluctuations only affect the isoscalar density, $\rho_p(\vec{r}) = y_p \rho_p(\vec{r}) \;\; ; \;\; \delta\rho_p(\vec{r}) = y_p \delta\rho_p(\vec{r})$, where the proton fraction y_p is assumed to be a constant. The free energy density variation induced by a finite size density fluctuation of wave number k and amplitude a is given by:

$$\delta f = a^2 \left(\frac{d\mu}{d\rho} + 2C_\rho k^2 + \frac{4\pi e^2 y_p^2}{k^2} \right) \; . \tag{1.1}$$

We consider for the initial homogeneous density ρ_0 a value lying inside the thermodynamic spinodal $d\mu/d\rho<0$, and a wave number k verifying the relation $\det C(k)<0$, where C is the curvature matrix, given in this simple one-dimensional case by the quantity inside parentheses in Eq.(1.1). The density evolution in time, following the initial fluctuation, is given by the Fick diffusion equation:

$$\frac{\partial^2 \delta\rho}{\partial t^2} = D \Delta \delta\rho \;\; , \tag{1.2}$$

where the diffusion coefficient is given by $D=dP/d\rho=d^2f/d\delta\rho^2$, and the initial condition is given by:

$$\delta\rho(\vec{r}, t=0) = a e^{i\vec{k}\cdot\vec{r}} + c.c. \tag{1.3}$$

At successive times the fluctuation is propagated in time according to:

$$\delta\rho(\vec{r}, t) = a e^{i(\vec{k}\cdot\vec{r} - \omega t)} + c.c. \tag{1.4}$$

Replacing in the Fick equation gives the *dispersion relation* :

$$\omega^2 \delta\rho = D k^2 \delta\rho \quad \Rightarrow \quad \omega = \sqrt{D} k \tag{1.5}$$

We can see that a negative diffusion coefficient leads to an imaginary frequence, that is (see (1.4)) a spontaneous amplification of the fluctuation in time. This spontaneous amplification of density fluctuation is the mechanism responsible of phase separation at the bulk limit. In the case of a finite nucleus, fluctuations cannot obviously amplify over macroscopic domains, but still they can be amplified over the length scale defined by the unstable wavelengths. This scale represents the spatial extension of the high density region, in a surrounding medium at the density corresponding to the dilute phase. We can interpret this phenomenon of *cluster formation* as the finite system counterpart of the bulk phase transition.

In such a situation it is clear that correlations among nucleons are essential and a mean-field description is completely inadequate. A complete correlated quantum theory of cluster formation is not presently available. However, it might be reasonable to assume that the inter-particle correlations are completed exhausted by cluster formation. In this hypothesis, the problem of correlated particles can be replaced by a much simpler problem of uncorrelated clusters. This assumption is at the heart of the Fisher model (Fisher, 1967) which was initially introduced in the field of condensed matter, and very successfully applied in the nuclear physics context from the early 80's.

2. The cluster model

The basic principle of a cluster model is the hypothesis that the inter-particle interactions and correlations lead to the formation of clusters. The problem of correlated particles is thus replaced by a problem of independent clusters, and all the complexity of the underlying physics is hidden in the energy functional of the clusters (*cluster self-energy*). As a first approximation, the vacuum cluster energy can be used. In this case we assume that the correlations are fully accounted by the clusterization. More sophisticated approaches, which are widely explored in the literature but we do not treat in these courses, consist in replacing the vacuum energy with an effective energy issued of a many-body calculation which takes into account the residual inter-cluster interactions.

Cluster models are used in a huge number of applications in condensed matter and astrophysics. In nuclear physics, the most recent applications concern multi-fragmentation, and the alpha-cluster structure of even-even nuclei. Here we limit ourselves to the formulation by Fisher, proposed to describe nucleation phenomena in the liquid-gas phase transition of simple fluids.

We consider a system of A nucleons constituted of n_a independent clusters of particle number a. The thermodynamic equilibrium in the grand-canonical ensemble is described by the partition sum:

$$Z_{\beta\mu} = Tr \exp - \beta\left(\hat{H} - \mu\hat{N}\right) = \sum_K \exp - \beta\left(E_K - \mu N_K\right) \tag{2.1}$$

where the sum extends over microstates, and E_K, N_K are the corresponding eigenvalues. In the ideal cluster system we are considering, these latter are given by :

$$\begin{aligned} N_K &= \sum_{a=1}^{A} n_a^{(K)} a \\ E_K &= \sum_{a=1}^{A} n_a^{(K)} e_a \end{aligned} \tag{2.2}$$

where e_a is the energy of a cluster of size a, and $n_a^{(K)}$ is its multiplicity (i.e., number of occurrences) in the state K. The partition sum is thus factorized :

$$Z_{\beta\mu} = \prod_{a=1}^{A} \sum_n \exp - \beta n\left(e_a - \mu a\right) \tag{2.3}$$

The cluster occupation numbers have to be specified. For a fermionic nuclear species, n=0,1:

$$Z_{\beta\mu}^F = \prod_{a=1}^{\infty}\left(1 + \exp - \beta\left(e_a - \mu a\right)\right) \tag{2.4}$$

If particles are bosons, the sum over occupations is a convergent (if $\mu < e_a/a$) geometric series giving :

$$Z_{\beta\mu}^B = \prod_{a=1}^{\infty}\left(1 - \exp - \beta\left(e_a - \mu a\right)\right)^{-1} \tag{2.5}$$

The classical limit, which is supposed to be valid at the low densities associated to our applications, consists in taking the limit $\beta\mu \to \infty$ such that

$\pm \ln\left(1 \pm \exp - \beta\left(e_a - \mu a\right)\right) \approx \exp - \beta\left(e_a - \mu a\right)$:

$$\ln Z_{\beta\mu} = \sum_{a=1}^{\infty} \exp - \beta n\left(e_a - \mu a\right) \tag{2.6}$$

The result is :

$$Z_{\beta\mu} = \prod_{a=1}^{\infty} \sum_{n=0}^{\infty} \frac{z_a^n}{n!} \quad ; \quad z_a = \exp - \beta\left(e_a - \mu a\right) \tag{2.7}$$

We can recognize a product of ideal gas partition sums. The extension to two types of particles (protons and neutrons) implies the introduction of two independent chemical potentials, $\mu = \left(\mu_n + \mu_p\right)/2, \quad \tilde{\mu} = \left(\mu_n - \mu_p\right)/2$. The partition sum immediately results:

$$Z_{\beta\mu\tilde{\mu}} = \prod_{n,z=1}^{\infty} \sum_{k=0}^{\infty} \frac{z_{nz}^k}{k!} \quad ; \quad z_{nz} = \exp - \beta\left(e_{nz} - \mu(n+z) - \tilde{\mu}(n-z)\right) \tag{2.8}$$

3. The cluster partition function

In the previous section we have considered that the cluster states are characterized by a=n+z, i=n-z, e=m(n,z) as unique quantum numbers. In our applications clusters are nothing but nuclei, meaning that they have an internal structure corresponding to different excited states of energy $e_{nz} = e_{nz}^0 + e_{nz}^*$, and the wave functions associated to their center-of-mass are plane waves, associated to linear momenta p_{nz}. The cluster partition sum becomes:

$$Z_{\beta\mu\tilde{\mu}} = \sum_{\{n_{nz}\}} \sum_{\{\vec{p}_{nz}\}} \sum_{\{e_{nz}^*\}} \exp -\beta\left(E_K - \mu A_K - \tilde{\mu} I_K\right) \quad , \tag{3.1}$$

where :

$$E_K = \sum_{n,z} n_{nz}^{(K)} \left(\frac{\vec{p}_{nz}^{\,2}}{2m_{nz}} + e_{nz}^0 + e_{nz}^* \right) \quad . \tag{3.2}$$

For each cluster, the sum over linear moment is given by the density of states of plane waves with periodic boundary conditions :

$$\sum_{\{\vec{p}_{nz}\}} = \frac{V}{h^3} \int d^3 p \quad , \tag{3.3}$$

and the sum over internal states can be written as integral by introducing the density of states :

$$\sum_{\{e_{nz}^*\}} = \int de \rho_{nz}(e) \quad ; \quad \rho_{nz}(e) = \sum_{i=1}^{\infty} \delta(e_i - e) \quad , \tag{3.4}$$

where the sum runs over the excited states of the nucleus (n,z). Replacing in Eq.(3.1) we get :

$$Z_{\beta\mu\tilde{\mu}} = \prod_{n,z} \sum_{n=0}^{\infty} \frac{V}{h^3} \int d^3 p \exp -\beta \left(\frac{\vec{p}_{nz}^{\,2}}{2m_{nz}} + e_{nz} - \mu a - \tilde{\mu} i \right) \int de \rho_{nz}(e) \exp -\beta e \quad . \tag{3.5}$$

We can see that the simple functional form $Z_{\beta\mu} = \prod_{a=1}^{\infty} \sum_{n=0}^{\infty} \frac{z_a^n}{n!}$ is preserved. This form comes from the statistical independence of clusters, i.e. the fact that we can write the total energy according to Eq.(2.2). In turn, this additivity is due to the absence of two-body operators in the Hamiltonian. The integral over linear momenta is a gaussian integral that can be solved analytically :

$$\int d^3 p \exp -\beta \frac{\vec{p}^{\,2}}{2m_{nz}} = \left(2\pi m_{nz} T\right)^{3/2} \quad . \tag{3.6}$$

The internal part is written as :

$$\int_0^{\infty} de \rho_{nz}(e) \exp -\beta e = g_{nz}^{\beta} \quad , \tag{3.7}$$

and gives a temperature dependence to the degeneracy of the states. The final result is:

$$
\begin{aligned}
Z_{\beta\mu\tilde{\mu}} &= \prod_{n,z=1}^{\infty}\sum_{n=0}^{\infty}\frac{z_{nz}^{n}}{n!} \\
z_{nz} &= \frac{V}{h^3} g_{nz}^{\beta}\left(2\pi m_{nz}T\right)^{3/2}\exp-\beta\left(e_{nz}-\mu_{nz}\right) \\
\mu_{nz} &= \mu(n+z)+\tilde{\mu}(n-z)
\end{aligned}
\quad . \qquad (3.8)
$$

The use of standard statistical tools allows obtaining all the observables. We hereby report the average cluster multiplicity, the relation between particle numbers and chemical potentials, as well as the pressure.

$$
\begin{aligned}
\langle n_{nz}\rangle &= \frac{\partial \ln Z_{\beta\mu\tilde{\mu}}}{\partial \beta\mu_{nz}} = z_{nz} \\
N_{tot} &= \frac{\partial \ln Z_{\beta\mu\tilde{\mu}}}{\partial \beta\mu_{n}} = \sum_{n,z}\langle n_{nz}\rangle n \\
Z_{tot} &= \frac{\partial \ln Z_{\beta\mu\tilde{\mu}}}{\partial \beta\mu_{p}} = \sum_{n,z}\langle n_{nz}\rangle z \\
\beta p &= \frac{\partial \ln Z_{\beta\mu\tilde{\mu}}}{\partial V} = \frac{1}{V}\sum_{n,z}\langle n_{nz}\rangle
\end{aligned}
\quad . \qquad (3.9)
$$

It is especially interesting to remark that we find back for the pressure the same functional form as for an ideal classical gas. The evaluation of the total energy requires the knowledge of the density of states. In the previous course we have demonstrated that, in the low excitation energy limit and in the framework of the independent particle model, the density of states reads :

$$\rho_{nz}(e) = C\exp 2\sqrt{aa_{nz}\left(e-e_{nz}^{0}\right)} \quad , \qquad (3.10)$$

where a_{nz} is the level density parameter. In the same approximations, the average energy at finite temperature is :

$$\langle e_{nz}\rangle_{\beta} = e_{nz}^{0} + a_{nz}aT^{2} \quad . \qquad (3.11)$$

The temperature dependent degeneracy can be modelized in the saddle point approximation, using again the low temperature approximations :

$$g_{nz}^{\beta} = \int_{0}^{\infty} de\exp-\beta(e-Ts_{nz}(e)) \cong \exp-\beta < (e-Ts_{nz}(e)) >_{\beta} = \exp a_{nz}aT \quad . \qquad (3.12)$$

This allows calculating the functional relation between energy and temperature :

$$E_{tot} = -\frac{\partial \ln Z_{\beta\mu\tilde{\mu}}}{\partial \beta\mu} = \sum_{n,z}\langle n_{nz}\rangle\left(e_{nz}^{0}+\frac{3}{2}T+a_{nz}aT^{2}\right) \qquad (3.13)$$

4. Link with the bulk phase transition

The cluster model allows predicting the fragment distribution issued of the fragmentation of a highly excited nucleus, under the hypothesis that a good degree of equilibrium was reached in the collision (Bohr independence hypothesis). The most sophisticated versions of this model, which are formulated in the microcanonical ensemble and consider the Coulomb interactions among fragments, have been massively compared to experimental data during the last 20 years and have shown an excellent predictive power. An example is given in the figure below.

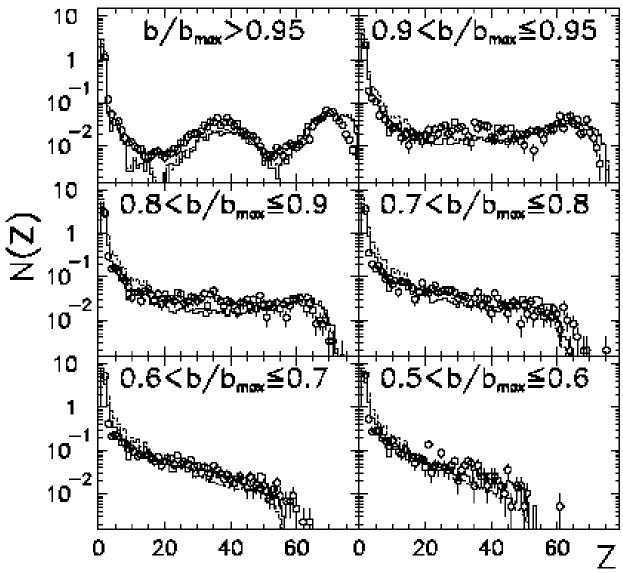

Fig. 4:
Mean elemental event multiplicity $N(Z)$ in different b/b_{max} intervals. The circles show experimental data and the solid/dashed histograms the results of SMM filtered/not filtered predictions.

We show in this section that the thermodynamic limit of this model contains a first order LG phase transition. This confirms again that clusterization can be considered as the finite system counterpart of a bulk LG phase transition. Indeed different physical situations exist where the phase transition is quenched. One such case is given by the finite size, which prevents macroscopic density fluctuations to emerge. Another interesting physical case, which we have explored in the last course, is the case of stellar matter. In this environment the presence of an incompressible electron background prevents the development of long range fluctuations even if we are at the thermodynamic limit. A characteristic of a system exhibiting a LG phase transition is that matter is not only unstable with respect to phase separation, but it is also unstable with respect to finite size density fluctuations. In the physical situations where the phase transition is quenched, it is therefore replaced by cluster formation.

In order to study the thermodynamic limit, the grand-canonical formulation is perfectly adequate and we will stick to it.

Let us consider again the expression of the multiplicity of clusters with a size a=n+z and isospin i=n-z predicted by the cluster model at a temperature $T=\beta^{-1}$ and chemical potentials $\mu,\tilde{\mu}$ (eq.(3.9)) :

$$\langle n_{ai}\rangle z_{nz} = \frac{V}{h^3}\left(2\pi m_{nz}T\right)^{3/2}\exp-\beta f_{ai} \quad , \tag{4.1}$$

with

$$f_{ai} = e_{ai} + \left\langle e^*_{ai}\right\rangle_\beta - T\left\langle s_{ai}\right\rangle_\beta - \mu a - \tilde{\mu} i \quad . \tag{4.2}$$

We can use a simplified semi-empirical mass formula for the ground state energy (y=i/a):

$$e_{ai} = \left(-c_V a + c_S a^{2/3}\right)\left(1 - c_I y^2\right) + \frac{c_C}{4}\left(1-y\right)^2 a^{5/3} \quad . \tag{4.3}$$

In order to have a good predictive power it is important to add to ph excitations also collective states, which correspond to rotations and vibrations of the nuclear surface (see course 2). To this aim, it is customary to introduce an effective entropic surface term proportional to $a^{2/3}$

$$g^\beta_{ai} = \exp\left(a_{nz}Ta + c_\beta a^{2/3}\right) \quad .. \tag{4.4}$$

Equation (4.2) becomes :

$$f_{ai} = -\left(c_V - a_0 T^2 + \mu + \tilde{\mu} y\right)a + \frac{c_C}{4}\left(1-y\right)^2 a^{5/3} + c^\beta_s\left(1 - c_I y^2\right)a^{2/3} \quad , \tag{4.5}$$

where $c^\beta_s = c_S - c_\beta$ is a decreasing function of temperature, which goes to zero at a temperature $T=T_c$. The presence of a dense liquid phase is linked in this model to the emergence of clusters of diverging size. The probability of an infinite or percolating cluster depends on the sign of the function f in the limit $a \to \infty$. We can see from Eq.(4.5) that, if $T=T_C$, the cluster size distribution is an increasing (decreasing) exponential function for

$$\mu > (<)\mu_C = -c_V + a_0 T_c^2 , \tag{4.6}$$

which signals the transition from liquid to gas. For $T=T_C$ et $\mu=\mu_C$, the distribution is independent of the cluster size : all size fluctuations are equally probable, in agreement with the physical expectation at a critical point.

At a temperature lower than the critical temperature, Eq.(4.6) still defines the transition point between the liquid and gas phase, but the two phases present a surface tension allowing the existence of intermediate size clusters at the interface.

Bibliography

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Exercices

1. Consider the Fisher model described by the grand-canonical partition sum :
$Z_{\beta\mu} = \prod_{a=1}^{\infty} \sum_{n=0}^{\infty} \frac{z_a^n}{n!}$; $z_a = \frac{V}{h^3}(2\pi m_a T)^{3/2} \exp -\beta(f_a - \mu a)$ avec $f_a = \langle e_a \rangle_\beta - T \langle s_a \rangle_\beta$,
$\langle e_a \rangle_\beta = -c_V a + c_S a^{2/3} + a_0 T^2 a$; $\langle s_a \rangle_\beta = 2a_0 T^2 a + c_s T T_c^{-1} a^{2/3} - \tau \ln a$ where c_V, c_S, a_0, T_c, τ are all parameters >0.
   a. Show that the cluster size distribution exhibits a power law for $T = T_c \quad \mu = c_V - a_0 T_c^2$ (critical point). We will use τ=4 in the following.
   b. Show that the baryon density is a finite number at the critical point.
   c. Show that the susceptibility $\chi = \frac{\beta}{V} \frac{\partial^2 \ln Z_{\beta\mu}}{\partial(\beta\mu)^2}$ diverges at the critical point.
   d. Show that the specific heat $C = \frac{\beta^2}{V} \frac{\partial^2 \ln Z_{\beta\mu}}{\partial\beta^2}$ diverges at the critical point.