Equilibrium and non-equilibrium multifragmentation

C. O. Dorso¹, A. Chernomoretz¹, and J. A. López²

¹ Facultad de Ciencias Exactas y Naturales, Universidad de Buenos Aires, ZIP-1428 Ciudad Universitaria, Pabellón I, Buenos Aires, Argentina
² Department of Physics, University of Texas at El Paso, El Paso, Texas, 79968 U.S.A.

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Abstract. In this work we report results obtained for fully microscopic calculation of space constrained and unconstrained highly excited finite systems. We analyze the properties of the caloric curves (CC) for both systems. We also study the behavior of thermal response functions (TRF), and show that under certain conditions they attain negative values. In order to get a better understanding of these features we explore the behavior of the fluctuations of the kinetic energy and its relation to fragment spectra.

1. Introduction

The possibility of getting information about the thermodynamics of nuclear matter from the analysis of intermediate energy heavy ion collisions has triggered a lot of interest in this field [1, 2]. One of the most challenging features that nuclear multi-fragmentation phenomena presents, is that signals of phase transitions of different orders can be extracted from experimental data. On one hand signatures of first order phase transition have been reported [3–6]. On the other, critical behavior have also been obtained assuming Fisher-like scaling relations for fragment distributions [7–10]. Moreover, in recent works [11, 12] is suggested that the observed critical behavior is compatible with a first order phase transition, and is a property of finite systems.

Several statistical descriptions of the nuclear multi-fragmentation process, e.g. the statistical multifragmentation model (SMM) [13] and the microcanonical Metropolis Monte Carlo model (MMMC) [4], employ the concept of a freeze-out volume. This hypothesis relies on the assumption that inside of a given volume an ensemble of equilibrated fragments exists. However, from the experience gained in numerical simulations, such a concept as freeze-out volume, even as an approximation, does not seem to be completely correct. It is then important to study which kind of differences arise as a consequence of not assuming a finite volume scenario, in which
equilibration is attained, as it is not a priori evident that the evolution of a fragmenting system confined in a finite volume would produce the same macroscopic observable when compared with a non confined one.

In previous works [16,17] we have studied the fragmentation of a simple classical system where the dynamics is governed by a Hamiltonian with a two body interaction Lennard-Jones term. A microscopic description, employing molecular dynamics techniques, was used in order to adequately handle the possible presence of a non equilibrium behavior. It was shown that a fragmentation time can be defined, after which a certain degree of local equilibrium is achieved in the system. This fact allowed us to calculate a caloric curve, defined as the temperature of the system at fragmentation time, for our expanding-fragmenting system, which is characterized by the absence of a vapor branch.

The aim of the present communication is to study how the restriction of a finite volume, and the corresponding imposition of equilibration, affects some of the results obtained in the unconstrained case.

This paper is organized as follows. In Section I) we will describe the model used in our simulations of unconstrained and constrained systems. In Section II) we study the caloric curves (CC) for both cases. In Section III) we study the Thermal Response functions for both cases, and in Section IV) we focus on the behavior of kinetic energy fluctuations. Finally conclusions are stated.

2. Numerical simulations

The system under study is composed by excited drops made up of particles interacting via a 6-12 Lennard Jones potential, which reads:

$$V(r) = \begin{cases} 
4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right] & r \leq r_c \\
0 & r > r_c 
\end{cases} \quad (1)$$

We took the cut-off radius as $r_c = 3\sigma$. Energies and distances are measured in units of the potential well ($\epsilon$) and the distance at which the potential changes sign ($\sigma$), respectively. The unit of time used is: $t_0 = \sqrt{\sigma^2 m / 48\epsilon}$. We integrated the set of classical equations of motion using the well known Verlet algorithm [14]. Initial conditions were constructed using the already presented method of cutting spherical drops composed of 147 particles out of equilibrated and periodic 512 particles per cell L.J. system [17].

2.1. Unconstrained expanding System

In this section we summarize the main results obtained in previous work for the non-constrained fragmenting system (see [17] for details).

One of the key ingredients in a fully microscopic dynamical analysis of the fragmentation process is the determination of the time at which fragments are
formed. In order to detect the fragments we use the MST and ECRA fragment recognition algorithms. In the MST algorithm clusters are defined according to: a particle $i$ belongs to a cluster $C$ if there is another particle $j$ that belongs to $C$ and $|r_i - r_j| \leq r_{cl}$, where $r_{cl}$ is a parameter called clusterization radius (In this work we used $r_{cl} = r_{cut} = 3\sigma$). On the other hand, in [18] it was shown that a different recognition method, the Early Cluster Formation Model and its companion practical realization the so called Early Cluster Recognition Algorithm (ECRA), outperforms the MST clusterization algorithm in the sense that it finds the asymptotic fragmentation pattern at early stages in the evolution, when fragments are still not observables in configuration space. At that time the system still looks like a rather compact piece of excited matter. The ECRA fragments are associated with the set of clusters $\{C_i\}$ for which the sum of the fragment internal energies attains its minimum value:

$$
\{C_i\} = \text{min}\{C_i\}[E_{\{C_i\}} = \sum_i E^c_{int}]
$$

$$
E^c_{int} = \sum_i \left[ \sum_{j \in C_i} K^{c,m.}_j + \sum_{j,k \in C_i, j \leq k} V_{j,k} \right]
$$

where the first sum in (2) is over the clusters of the partition, $K^{c,m.}_j$ is the kinetic energy of particle $j$ measured in the center of mass frame of the cluster which contains particle $j$, and $V_{ij}$ stands for the inter-particle potential. We dub the partition found by ECRA as the most bound density fluctuation in phase space (MBDF) and we define as time of fragment formation ($\tau_{ff}$) the time at which the MBDF attain microscopic stability. In this way $\tau_{ff}$ is related to the time at which the systems switches from a regime dominated by fragmentation to one dominated by evaporation of light aggregates (mostly single particles) by the excited fragments.

Once the time of fragment formation is determined it is possible to calculate several properties of the system at fragmentation time. For this purpose the expanding system is decomposed in concentric shells and the mean radial velocity is calculated. It is found that the expansion is almost linear with the distance to the center of mass of the system, and that a local temperature can be defined as the fluctuations of the velocity around the local expansive collective motion. Moreover, the isotropic character of those fluctuations supports the idea that local equilibrium is achieved [17]. Moreover, it has been found that at $\tau_{ff}$ most of the system is still interacting, and that the local temperature of the inner shells attain a rather constant value that can be consistently considered as the temperature of the system at fragmentation time.

2.2. Constrained System

In order to study the consequences of imposing a finite volume constraint to our system we used a spherical confining “wall”. The considered external potential behaves like $V_{wall} \sim (r - r_{wall})^{-12}$ with a cut off distance $r_{cut} = 1\sigma$, where it smoothly became zero along with its first derivative.
Inside this potential, a highly excited drop was initialized as described above and the corresponding equations of motion were integrated.

In this case the standard prescription for temperature calculation in the microcanonical ensemble was used, i.e. the kinetic energy ($K$) was related to the temperature of our $N$-particles system using:

$$T = \frac{2}{3(N-1)} K$$

3. Caloric curves

One of the thermodynamics quantities that remains useful for small systems is the caloric curve, i.e. the functional relationship of the system temperature with its excitation energy.

3.1. Unconstrained expanding System

For the unconstrained system the caloric curve is defined as the functional relationship between the temperature of the system and its excitation energy at fragmentation time i.e at $\tau_{ff}$. The resulting caloric curve (CC) for the unconstrained system is displayed in Fig. 1 a). In this figure circles denote the temperature of the system, as a function of the energy. On the same figure a curve denoted by squares shows the value of the total kinetic energy, i.e. including both the fluctuations around the collective motion and the collective motion itself. From the figure it is then obvious that collective motion begins to be noticeable around a value of $E = 0$, and it becomes dominant at around $E = 2\epsilon$. Two main features are to be noticed, in first place the CC develops a maximum, and second the CC has no “vapor branch” but develops a rather constant behavior. This constant behavior is related to the presence of a radial collective motion that behaves as a heat sink.

3.2. Constrained System

In this case we need to calculate the temperature of an equilibrated system, which is given by $T = \frac{2}{3(N-1)} K$

In Fig.2 a) we show the behavior of $T(E, \rho)$ for three representative density values. For all density values a vapor branch is clearly observed. This is a direct consequence of the presence of the wall, which imposes equilibration.

The effect of the boundaries on the CC is not important as long as the total energy in the system is small enough, and the expansive collective motion (in the unconstrained case) can be neglected. But this is not the case for highly excited finite systems, for which the presence of the constraining wall prohibits the formation of the expansive radial motion. For that range of energies the local equilibrium
features found in the fragmentation of the unconstrained system are replaced by global.

It is also seen that as the density decreases a loop in the CC develops.

4. Thermal response function

In recent works [4,6,15] a lot of attention has been paid to the role played by the behavior of the specific heat (or more generally speaking: the thermal response function, TRF) as a signal of the occurrence of a phase transition in finite systems. Moreover, it has been shown (see [4]) that the presence of a negative branch in the TRF can be related to a first order phase transition taking place in an isolated finite non extensive system.

4.1. Unconstrained expanding System

The calculation of the TRF can straightforwardly be performed for the unconstrained system using the already calculated CC and taking into account that the respective TRF’s is given by:

\[ TRF = \left( \frac{\partial T}{\partial E} \right)^{-1} \]  

In Fig.1b) we show the TRF for the unconstrained system. Two poles can be seen. The first one \((E \sim -0.5 \epsilon)\) signals the entrance of the system into the multifragmentation regime, while the second one is related to the leveling off in the corresponding CC, and can be related to the increasing limit imposed by the strong flux to the “thermalization” of the total available energy.

4.2. Constrained System

We now analyze the constrained system and try to relate its behavior to the one found for the unconstrained case. As was already mentioned, due to the intrinsic non-equilibrium character of the fragmentation process in the non-confined case, an assumption of local equilibrium instead of a global one has been found to be more appropriate. Accordingly, distributions of density and temperature, and not unique values, are necessary to described the system at fragmentation time in a rigorous way (see [17]). Two different criteria can be adopted in order to choose the appropriate volume for the constrained case (which we will call freeze-out volume). On one hand at \(\tau_{ff}\) the local density value for the inner cells (where most of the mass is present at the time of fragment formation) remains rather constant \((\rho \sim 0.08 \sigma^{-3})\) for a broad energy range [17]. Accordingly, one can choose \(V_{fo}\) in order to attain such density value in the constrained case. This choice corresponds to \(r_{wall} \sim 8 \sigma\) and a freeze out density value of \(\rho_{fo} \sim \rho_0/10\) .

On the other hand a different approach can be adopted by matching the temperature of the free expanding system, at the onset of fragmentation, with the tem-
perature of the constrained case at the transition region. This can be accomplished at a density value of $\rho_{fo} \sim 0.02\sigma^{-3} \sim \rho_0/40$.

In Fig. 2b) the corresponding TRF for the constrained case is shown. We include the curves for $\rho_{+fo}$ and $\rho_{-fo}$, and an intermediate $\rho^0 \sim \rho_0/20$ value ($\rho_{-fo} < \rho^0 < \rho_{+fo}$). We notice that two poles and a negative branch can be observed for the $\rho_{-fo}$ TRF curve. In this case we can relate the first pole with the onset of the transition, and the second one with the entrance in the gas phase. For $\rho = \rho^0$ it can be seen that the curve exhibits a qualitatively similar behavior, the distance between the two poles is just reduced. Nevertheless, for higher density cases, such as $\rho_{+fo}$, the two poles merge into a single “singularity” limited by finite-size effects. In that case, the TRF remains positive for all energies, showing a peak as a signature of the transition, as a consequence of the fact that the corresponding caloric curve does not display a loop but instead a change of slope. In brief, the systems behaves as undergoing a first order phase transition at low densities and behaves as undergoing a second order one in the high density case.

5. Fluctuations

In order to further explore the thermodynamic behavior of our systems we use an approach introduced in [15], in which the behavior of the fluctuations in the kinetic energy is used as an indicator of the occurrence of a phase transition of first order in finite systems. In [15] it is shown that for an isolated and equilibrated system in which the total energy can be decomposed as: $E = E_1 + E_2$ the heat capacity can be calculated as:

$$C \approx \frac{C_1^2}{C_1 - \sigma_1^2/T^2}$$

where $C_1$ is the heat capacity associated with the subsystem-1, $\sigma_1$ is the fluctuation of the partial energy $E_1$, and $T$ the temperature of the system. Consequently, the presence of poles and negative values in TRF’s can be associated to abnormally large fluctuations of the partial energy stored in subsystem-1, i.e. $\sigma_1^2 \geq C_1 T^2$, during the phase transition.

5.1. Unconstrained System

We now try to apply the above mentioned analysis for the unconstrained system. In order to do that, we considered the central region (or core) defined by $r <= 6\sigma$, and we calculated the total energy, $E_{core}$, and the number of particles $N_{core}$ that remains inside the core at fragmentation time. Binning the whole set of events according to these variables we can sample unconstrained fragmenting events in a pseudo-microcanonical way in what concerns core-quantities at $\tau_{ff}$. In Fig. 3 we show the behavior of the relative fluctuation of the core kinetic energy, $A(E)$,
Equilibrium and non-equilibrium multifragmentation defined as:

\[ A(E) = N \frac{\sigma_k^2 |E|}{\langle K \rangle |E|^2} \]  

(5)

where we used, \( K \equiv K_{\text{core}} \), \( E \equiv E_{\text{core}} \), \( N \equiv N_{\text{core}} = 60 \pm 5 \), and \( \langle \rangle \) stands for an average over the microcanonical sampling of configurations. In the same figure we include the canonical reference level. Even with a severe reduction of statistics, a region of large fluctuations in \( K_{\text{core}} \), for \( N_{\text{core}} \) and \( E_{\text{core}} \) fixed, can be recognized.

5.2. Constrained System

In Fig.4 a) we show the kinetic energy relative fluctuation, \( A(E) \), for the constrained case. Direct calculations of equation 5 for both, \( \rho^+_{f_0} \) and \( \rho^-_{f_0} \), are shown. The estimation of \( A(E) \) derived for the respective CC’s in Fig.4 is also included. A rather good agreement can be seen between both ways of calculating this magnitude. In the same graph a reference level marks the canonical value for \( A(E) \). As expected, for the appropriate range of energies, “unusually” large relative fluctuations for the \( \rho^-_{f_0} \) case can be observed, while for the \( \rho^+_{f_0} \) case a local maximum, that does not exceed the canonical value can be seen.

In order to further clarify this point we study the relation between the fluctuations and fragment distributions for this systems. We have analyzed the configurations obtained for the constrained systems with the MST and ECRA algorithms. MST analysis is particular relevant for the study of TRFs because it shows the ability of the system to disassemble in a set of non interacting pieces of matter. As such the presence of MST clusters, smaller that the total mass of the system, can be associated with the formation of surfaces. On the other hand ECRA clusters will be sensible to the interparticle correlations in q-p space. ECRA clusters can be formed inside MST clusters and then reveal finner details of the state of the system.

In Fig.5 we show the fluctuations of the kinetic energy, the size of the maximum ECRA cluster and the size of the maximum MST cluster as a function of time for \( \rho^+_{f_0} \) and \( \rho^-_{f_0} \). (See figure captions for details). We first notice that for the \( \rho^+_{f_0} \) case the size of the maximum MST cluster is almost constant in time and comprises most of the mass of the system. In this case no surfaces are formed in the system, as such an average over time of the fluctuations in kinetic energy will involve an average over an ensemble of configurations in which a big drop is always present. On the other hand the fluctuations in kinetic energy are well correlated with the fluctuations in the size of the ECRA clusters which take place “inside” the big MST drop. Nevertheless this fluctuations are not big enough to produce negative TRF.

When we focus on the less dense case, \( \rho^-_{f_0} \), we notice that there are big fluctuations in the size of the biggest MST clusters. This means that the time average above mentioned will involve, in this case, very dissimilar configurations with drops of different sizes, from ones in which a big drop is present to ones in which medium size drops are dominant. As such we will be averaging fluctuations around mean values which also fluctuate strongly. This can be understood in the following way.

If we assume that the configurations can be classified in liquid like (a big drop is
present) and vapor like (medium and small fragments dominate) it is easy to see that for the first one the fluctuations in kinetic energy are around the mean value of the kinetic energy characteristic of this liquid-like state. For the second set, fluctuations are calculated with respect to the mean value of the kinetic energy characteristic of the vapor-like state. When fluctuations are calculated for the complete set of configurations fluctuations are boosted by the difference between the two mean values considered (the mean kinetic energy for the liquid like states is bigger than the one for the vapor like states)

6. Conclusions

In this work we have presented a complete analysis of Caloric Curves, Thermal Response Functions for constrained and unconstrained excited liquid drops. We have shown that the effect of this kind of constraints is quite important, and that its main effect is to force the system to reach thermal equilibrium, and the appearance of a vapor branch in the Caloric Curves for constrained cases. This is an “equilibrium effect” that is not present for unconstrained systems.

It is interesting to notice that for the constrained case, as the system density is increased, the expected signals of a first order phase transition in the CC, TRF and kinetic energy fluctuations are gradually smoothed, and eventually disappear. This can be associated to the fact that at low densities “internal surfaces” can be developed in the constrained system allowing the transition to be traced in configurational space (i.e. MST clusters are formed reflecting the fact that well separated aggregates appear in coordinate space). Such a feature is not possible in the dense case, all of the time the system is composed by a big configurational cluster that comprises more than 95% of the total mass. Moreover for the constrained case, a relation between the fluctuations in size of the maximum ECRA-fragment and the kinetic energy fluctuations was established.

As a final remark we want to emphasize that, even some similarities can be found between the behavior of low density constrained systems and unconstrained ones at low energies, such a comparison fails at and above the energy that corresponds to the onset of the fragmentation process, i.e. when the collective radial modes begin to drive the evolution of the system.

As we mentioned, the role of this collective motion is to behave as a heat sink, precluding the system from developing a vapor branch, and freezing the most bound density fluctuations in phase space allowing them to become the asymptotic clusters. This is why a “local” equilibrium picture, and not a “global” one, is necessary to describe the fragmentation process correctly, i.e. taking into account the effects of the radial flux. This teaches us that when dealing with fragmentation phenomena of the kind appearing in nuclear multifragmentation, caloric curves which display vapor branches should be taken with caution, and consequently the results obtained from models that display such a feature should be at least critically reexamined.
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References

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Fig. 1. In figure (a): Caloric curve calculated for the expanding system (solid circles). The empty squares correspond to an estimation of a ‘fake-temperature’ that does not take into account in a proper way the collective motion and is calculated simply as a fraction of the total kinetic energy (see text for details). Figure (b) shows the associated thermal response function.

Fig. 2. Caloric curve for the constrained system, figure (a), and the respective thermal response functions, figure (b), for three different densities: \( \rho = \rho_{f_0}, \rho_{f_0}^0, \) and \( \rho_{f_0}^+ \) (circle, triangle, square symbols in figure (a), and full, dashed, dotted-dashed lines in figure (b), respectively).

Fig. 3. Relative kinetic fluctuation, \( A(E) \), for the constrained case calculated for different densities, as a function of the total energy. The symbols correspond to direct calculations for \( \rho = \rho_{f_0} \) (empty squares) and for \( \rho = \rho_{f_0} ( \text{solid circles}) \). The lines correspond to \( A \) estimations using the caloric curve (solid-line for \( \rho = \rho_{f_0} \), and dashed-line for \( \rho = \rho_{f_0}^+ \)). The dashed-dotted line shows the canonical expectation value for \( A \).
**Fig. 4.** Relative kinetic energy fluctuation, for the non-constrained system, as a function of the core-energy. The calculation includes only events with $N_{\text{core}} = 60 \pm 5$ particles inside the core.

**Fig. 5.** In figures labeled (a), the system kinetic energy is shown as a function of time for the constrained case. The temporal dependence of the mass of the biggest ECRA and MST clusters are shown in figures labeled (b) and (c) respectively. The upper panel corresponds to a density value equal to $\rho_f^0$, while the lower panel to $\rho_{fo}$.