First order phase transitions: equivalence between bimodalities and the Yang-Lee theorem

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First order phase transitions in finite systems can be defined through the bimodality of the distribution of the order parameter. This definition is equivalent to the one based on the inverted curvature of the thermodynamic potential. Moreover we show that it is in a one to one correspondence with the Yang Lee theorem in the thermodynamic limit. Bimodality is a necessary and sufficient condition for zeroes of the partition sum in the control intensive variable complex plane to be distributed on a line perpendicular to the real axis with a uniform density, scaling like the number of particles.

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Phase transitions are universal phenomena which have been theoretically understood at the thermodynamic limit of infinite systems as anomalies in the associated equation of state (EoS). They have been classified according to the degree of non-analyticity of the thermodynamic potential at the transition point. For finite systems, it is generally believed that phase transitions cannot be defined since thermodynamic potentials of a finite system in a finite volume are analytic functions. This situation can be thought as unsatisfactory since on one hand the thermodynamic limit does not exist in nature, and on the other hand many experimental efforts are devoted to understand the link between mesoscopic and macroscopic systems, especially for spectacular properties like phase transitions \[^{1}\].

A classification scheme valid for finite systems has been proposed by Grossmann \[^{2}\] using the distribution of zeroes of the canonical partition sum in the complex temperature plane. Alternatively, it has been claimed that first order phase transitions in finite systems can be related to a negative microcanonical heat capacity \[^{3}\] \[^{4}\] or more generally to an inverted curvature of the thermodynamic potential as a function of an observable which
can then be seen as an order parameter [9]. These anomalies can also be connected to the general topology of the potential energy surface [3]. These very different approaches share the common viewpoint that at the mesoscopic or microscopic scale phase transitions are not smeared to become loosely defined smooth state changes, but can be analyzed in a rigorous way without explicitly addressing the thermodynamic limit. If this is certainly important to progress in the experimental study of phase transitions in finite systems, it is also true that the connection between these mesoscopic phenomena and the thermodynamics of the macroscopic world is not clear. This is especially annoying since the definitions of phase transitions in finite systems are not in general equivalent and different conclusions can be drawn out of the observation of a physical phenomenon depending on the theoretical framework used. In particular it can be shown [7] that any generic channel opening may lead to a convexity anomaly of the microcanonical entropy which does not necessarily survive to the bulk. In the same way the distribution of zeroes close to the real axis may suggest a first order phase transition even for systems where the transition temperature diverges in the thermodynamic limit [8]. In order to have a coherent picture of the physical meaning of a phase transition in a finite system it is then important to analyze in detail the conditions of persistency of the signals towards the bulk and to make a clear bridge between the various definitions.

We have recently proposed the possible bimodality of the probability distribution of observable quantities as a connection between these ideas [9]. In this letter we further demonstrate that this definition is consistent with the Yang Lee theorem [10], i.e. with the standard definition of first order phase transitions in the thermodynamic limit. Since our definition is a generalization of the previous ones [2, 3, 4], the conditions of compatibility of the different signals and criteria proposed to identify first order phase transitions in finite systems can be explicitly worked out.

Let us consider that for each event $i$ we can measure a set of $K$ independent observables, $b_k^{(i)}$, which form a space containing one possible order parameter. We can sort events according to the results of the measurements $\bar{b}^{(i)} \equiv \{b_k^{(i)}\}$ and thus define a probability distribution $P(\bar{b})$. In the absence of a phase transition $\log P(\bar{b})$ is expected to be concave. An abnormal (e.g. bimodal) behavior of $P(\bar{b})$ or a convexity anomaly signals a first order phase transition [9].

This definition does not imply an a priori knowledge of the thermodynamic potential,
i.e. of the properties of the specific equilibrium reached in the experimental situation, but only requires the measurement of the event distribution. Moreover, it does not assume the a-priori knowledge of an order parameter; the order parameter can be extracted from the data themselves by looking at the best direction in the observable space to split the event cloud into two components. This topological definition is therefore easy to experimentally implement, and in fact it has already allowed to successfully recognize phase transitions in metallic clusters [11] and in the multifragmentation of atomic nuclei [12].

Moreover this definition has a wider application domain than the previous ones [2, 3, 4]; it can be used out of canonical or microcanonical equilibria, for open systems [9] and non-extensive statistics such as Tsallis equilibria [13]. In particular it can be viewed as an extension of the definition based on the convexity anomaly of the microcanonical entropy [3, 4] or more generally of any thermodynamical Gibbs potential [5]. Let us first discuss Boltzmann-Gibbs equilibrium which is obtained by maximizing the Shannon information entropy under the constraints of the various observables $b_k$ known in average. Then $P(\vec{b})$ can be written as

$$
\log P_{\vec{\lambda}}(\vec{b}) = \log \bar{W}(\vec{b}) - \sum_{k=1}^{K} \lambda_k b_k - \log Z_{\vec{\lambda}}
$$

where $Z_{\vec{\lambda}}$ is the partition sum of this intensive ensemble controlled by the Lagrange multipliers $\vec{\lambda}=\{\lambda^{(i)}_k\}$ and $\bar{W}(\vec{b}) = Z_0 P_0(\vec{b})$ is the partition sum of the statistical ensemble associated with fixed values $\vec{b}$ of all the observables. In the following we will call it the extensive ensemble using the word ”extensive” in the weak sense of ”observable” [14], meaning that observables are controlled on an event-by-event basis in the extensive ensemble. The two partition sums are related through the usual Laplace transform $Z_{\vec{\lambda}} = \int d\vec{b} \bar{W}(\vec{b}) \exp(-\vec{\lambda} \cdot \vec{b})$. Eq. (1) clearly demonstrates that the convexity anomalies of the thermodynamical potential $\log \bar{W}$ can be traced back from $\log P_{\vec{\lambda}}(\vec{b})$. Indeed, the equation of states of the extensive ensemble can be related to the derivative of $\log P_{\vec{\lambda}}(\vec{b})$

$$
\vec{\lambda}_{k}(\vec{b}) \equiv \frac{\partial \log \bar{W}(\vec{b})}{\partial b_k} = \frac{\partial \log P_{\vec{\lambda}}(\vec{b})}{\partial b_k} + \lambda_k.
$$

while the curvature matrix of $\log \bar{W}$ is the one of $\log P_{\vec{\lambda}}(\vec{b})$

$$
C_{kk'}(\vec{b}) \equiv \frac{\partial \lambda_{k}(\vec{b})}{\partial b_{k'}} = \frac{\partial^2 \log \bar{W}(\vec{b})}{\partial b_k \partial b_{k'}} = \frac{\partial^2 \log P_{\vec{\lambda}}(\vec{b})}{\partial b_k \partial b_{k'}}.
$$

It should be noticed that these relations are valid for every set of Lagrange multipliers $\vec{\lambda}$.
because in a finite system the probability distribution covers the entire accessible space of $\vec{b}$ and so contains the whole thermodynamics of the extensive ensemble. The only limitation can be a practical one for experiments or simulations: to accumulate enough statistics at every location.

Let us consider the simplified case of a unique direction $b_k \equiv b$ and so a unique Lagrange multiplier $\lambda_k \equiv \lambda$. If $\bar{W} (b)$ has an abnormal curvature, then the EOS $\bar{\lambda} (b) = \partial_b \log \bar{W} (b)$ presents a back-bending. For this extensive ensemble where $b$ is the control parameter, coexistence can be defined as the region where $\bar{\lambda}$ is associated to three values of $b$ because of the anomalous curvature. The definition of a first order phase transition as the occurrence of a negative heat capacity \cite{3, 4} is readily obtained if one identifies $S = \log \bar{W}$ as the microcanonical entropy, $b$ as the energy and $\lambda = \beta$ the inverse canonical temperature. The generalization to the EOS of any thermodynamic potential involving an extensive variable $b$ \cite{5} is also obvious. For example, a bimodal grand-canonical number of particle distribution is equivalent to a negative chemical susceptibility in the canonical ensemble, a bimodal distribution of magnetization to a negative magnetic susceptibility in the constant magnetization ensemble, a bimodal volume distribution in the isobar ensemble to a negative compressibility of the isochore ensemble, ...

For $\lambda$ in the region of anomalous curvature the associated probability distribution presents two maxima and a minimum. In the intensive ensemble eq.(1) where $\lambda$ is controlled, coexistence is then signalled by the bimodality of the probability distribution and the value of $\lambda$ for which the two maxima have equal height is the first order transition point, analogous to the usual Maxwell construction. Therefore, the definitions of a first order phase transition through the occurrence of an abnormal curvature of the thermodynamical potential of the extensive ensemble, or through the presence of a bimodal event distribution in the associated intensive ensemble are strictly equivalent.

Let us now consider the definition \cite{2} based on the zeroes of $Z_{\vec{\lambda}}$, the partition sum of the intensive ensemble eq.(1), in the complex $\vec{\lambda}$ plane. We shall now work out the relations between these zeroes and the concavity of $P_{\vec{\lambda}}$. Let us continue for simplicity with a single observable $b_k \equiv b$ and the associated Lagrange multiplier $\lambda_k \equiv \lambda$. Looking at the zeros of $Z_{\lambda}$ in the complex $\lambda$ plane, we first show that a bimodal distribution corresponds to a partition sum fulfilling the Yang Lee theorem in the double saddle point approximation \cite{9}. Then we will demonstrate the reciprocal: a distribution of zeroes which fulfills the Yang Lee theorem
is associated to a bimodal probability distribution for $b$.

Using equation (1) we see that the partition sum for a complex parameter $\gamma = \lambda + i\eta$ is nothing but the Laplace transform of the probability distribution $P_{\lambda_0}(b)$ for any arbitrary parameter $\lambda_0$.

$$Z_\gamma = \int db Z_{\lambda_0} P_{\lambda_0}(b) e^{-(\gamma - \lambda_0)b} \equiv \int db p_{\lambda_0}(b) e^{-\tilde{\gamma}b}$$

where $\tilde{\gamma} = \gamma - \lambda_0$ and we have defined $p_\lambda(b) = Z_\lambda P_\lambda(b)$. We can also write the partition sum at a complex point $\gamma = \lambda + i\eta$ as a Fourier transform of the probability distribution $p_\lambda(b)$

$$Z_\gamma = \int db p_\lambda(b) e^{-i\eta b}$$

If $P_\lambda(b)$ is monomodal we can use a saddle point approximation around the maximum $b_\lambda$ giving $Z_\gamma = e^{\phi_\gamma(b_\lambda)}$, with

$$\phi_\gamma(b) = \log p_\lambda(b) - i\eta b + \frac{1}{2} \eta^2 \sigma^2(b) + \frac{1}{2} \log (2\pi\sigma^2(b))$$

where $\sigma^{-2} = \partial^2_b \log p_{\lambda_0}(b)$. However, if $\tilde{W}_{\lambda_0}(b)$ has a curvature anomaly it exists a range of $\lambda$ for which the equation $\partial_b \log \tilde{W}_{\lambda_0} = \lambda - \lambda_0$ has three solutions, one minimum and two maxima $b^{(1)}_\lambda$ and $b^{(2)}_\lambda$. Let us first split the probability distribution into two normal components

$$p_\lambda(b) = p^{(1)}_\lambda(b) + p^{(2)}_\lambda(b)$$

$p^{(i)}_\lambda(b)$ being peaked at $b^{(i)}_\lambda$. The partition sum reads

$$Z_\gamma = e^{\phi^{(1)}_\gamma} + e^{\phi^{(2)}_\gamma} = 2e^{\phi^+\gamma} \cosh(\phi^-\gamma)$$

where $2\phi^+\gamma = \phi^{(1)}_\gamma + \phi^{(2)}_\gamma$ and $2\phi^-\gamma = \phi^{(1)}_\gamma - \phi^{(2)}_\gamma$. We can now use a double saddle point approximation around the two maxima which will be valid close to thermodynamical limit and we get $\phi^{(i)}_\gamma = \phi_\gamma(b^{(i)}_\lambda)$ according to eq. (6). The zeros of $Z_\gamma$ then correspond to

$$2\phi^-\gamma = i(2n + 1)\pi$$

The imaginary part is given by $\eta = (2n + 1)\pi/\Delta b$ where $\Delta b = b^{(2)}_\lambda - b^{(1)}_\lambda$ is the jump in $b$ between the two phases. For the real part we should solve the equation $\Re(\phi^-_\gamma) = 0$. In particular, close to the real axis this equation defines a $\lambda$ which can be taken as $\lambda_0$. If the bimodal structure persists when the number of particles goes to infinity, the loci of zeros
corresponds to a line perpendicular to the real axis with a uniform distribution as expected for a first order phase transition [10].

Let us now work out the necessary condition and show that a uniform distribution of zeroes perpendicular to the real axis with a density linearly increasing with the number of particles $N$ implies a bimodal probability distribution. Let us denote zeros as

$$\gamma_n = \lambda_0 + i(2n + 1)\frac{\pi}{N\delta} \quad \text{(10)}$$

such that the interval $2\pi/\delta$ contains $N$ uniformly distributed zeroes in agreement with the unit circle theorem [10]. Since all the zeros of $Z_\gamma$ are periodically distributed one can define an analytic function $f(\gamma)$ such that

$$Z_\gamma = 2 \cosh \left( (\gamma - \lambda_0) \frac{N\delta}{2} \right) f(\gamma) \quad \text{(11)}$$

Indeed since $Z_\gamma$ is analytic, $f(\gamma)$ could in principle diverge only on the zeroes of $Z_\gamma$; in these points however eq.(11) shows that $f$ is proportional to $\partial_\gamma Z_\gamma$, i.e. it is analytic. On the line of zeroes, relation (11) reduces to $Z_{\lambda_0+i\eta} = 2 \cos (\eta N\delta/2) f(\lambda_0 + i\eta)$. Using the fact that the partition sum along the line of zeroes is the Fourier transform of the reduced probability distribution at the transition point $\lambda_0$ (eq.(3), we can use the inverse Fourier transform to get the distribution $p_{\lambda_0}(b)$ at the transition point

$$p_{\lambda_0}(b) = \frac{1}{2\pi} \int d\eta Z_{\lambda_0+i\eta} e^{i\eta b} \quad \text{(12)}$$

Equation (12) can then be rewritten as

$$p_{\lambda_0}(b) = g_{\lambda_0} \left( b + \frac{N\delta}{2} \right) + g_{\lambda_0} \left( b - \frac{N\delta}{2} \right) \quad \text{(13)}$$

where

$$g_{\lambda}(b) = \frac{1}{2\pi} \int d\eta f(\gamma) e^{i\eta b}$$

is a distribution. Indeed if we compute $p_{\lambda}(b)$ a little above $\lambda_0$, $\lambda = \lambda_0 + \epsilon$ we get

$$p_{\lambda_0+\epsilon}(b) = e^{\epsilon N\delta/2} g_{\lambda_0+\epsilon} \left( b + \frac{N\delta}{2} \right) + e^{-\epsilon N\delta/2} g_{\lambda_0+\epsilon} \left( b - \frac{N\delta}{2} \right) \quad \text{(14)}$$

For large $N$ only the first term survives

$$p_{\lambda_0+\epsilon}(b) \simeq e^{\epsilon N\delta/2} g_{\lambda_0+\epsilon} \left( b + \frac{N\delta}{2} \right) \equiv p^{(1)}_{\lambda_0+\epsilon}(b) \quad \text{(15)}$$
which is the first term in the distribution at the transition point eq.(12). If we conversely compute $p_\lambda(b)$ a little below the transition point we get

$$p_{\lambda_0 - \varepsilon}(b) \simeq e^{N\delta/2} g_{\lambda_0 - \varepsilon} \left( b - \frac{N\delta}{2} \right) \equiv p_{\lambda_0 - \varepsilon}^{(2)}(b)$$

(16)

which is the second term in the transition point distribution eq.(12). At the transition point $p_{\lambda_0}(b)$ is the sum of two shifted (identical) distributions, $p_{\lambda_0}^{(1)}$ and $p_{\lambda_0}^{(2)}$. If $g_\lambda$ is a normal (i.e. monomodal) distribution the central limit theorem guarantees that for a large number of particles its width will scale as $\sqrt{N}$, i.e. will grow slower than the distance between the two peaks that scales as $N$. This implies that (for not too small $N$) $p_{\lambda_0}(b)$ is bimodal. This stays true if $g_\lambda$ itself is bimodal (or multimodal), with the only difference that the total distribution $p_{\lambda_0}(b)$ will then represent the coexistence between more than two phases. Let consider the simplest case of a bimodal structure for $p_{\lambda_0}(b)$, close to the thermodynamical limit; a little before the transition point only the lowest peak of the $b$ distribution is present, while a little above only the second one remains. At the transition both are present and the most probable $b$ jumps from $b_\prec = \bar{b} + N\delta/2$ to $b_\succ = \bar{b} - N\delta/2$ where $\bar{b}$ is the maximum of $g_{\lambda_0}(b)$. $\delta$ represents the discontinuity of the variable $b$ per particle. If $b$ is the energy $\delta$ is the latent heat per particle.

The important difference between finite systems and the thermodynamic limit is that in the latter case the probability distribution is bimodal only at the transition point $\lambda_0$. On the other hand in finite systems, in an interval $\Delta\lambda$ of non-zero measure around the transition point $\lambda_0$ the two phases coexist, i.e. the distribution is bimodal, each peak being associated with a phase having a finite probability of occurrence. Eq.(14) shows that this interval is the larger the smaller is $\delta$, i.e. the lower is the density of zeroes. This extension of the phase transition point to a region in which the intensive ensemble presents a bimodal distribution makes a direct measurement of phase coexistence possible [12], contrary to the common belief that phase transitions would only be loosely defined out of the thermodynamic limit. Moreover, the fact that the distribution is non zero between the two maxima $b_\prec$ and $b_\succ$ in the case of small systems (see eq.(14)) implies that microstates can be accessed that do not exist at the thermodynamic limit in the intensive ensemble. These states are specific of the coexistence region and can lead to spectacular phenomena as negative heat capacity [3] and negative compressibility [4] in the extensive ensemble.
This demonstration can be extended to account for more important finite size effects \[13\] when zeroes \(\gamma_n\) are still periodic but not yet uniformly distributed on a straight line \[2, 15\]. Indeed, it is important to remark that the distribution of zeroes is periodic in the imaginary direction before the thermodynamic limit. If for example we consider the grandcanonical ensemble, the Lagrange parameter \(\lambda \equiv -\alpha \equiv -\beta \mu\) represents the logarithm of the fugacity \(z = e^\alpha\) and the observable is the number of particles \(b \equiv N\). The partition sum \(Z_\alpha \propto e^{\alpha N}\) in the complex \(\alpha\) plane is periodic for any arbitrary value of \(N\). In the more general case, this will not be true for very small systems since the extensive variables \(b_k\) are not in general proportional to the number of constituents of the system. However, if the forces are short-ranged and \(N\) is sufficiently high, \(b_k \propto N\), which guarantees the periodicity of \(Z_\lambda\) in the complex \(\lambda\) plane. This periodicity is a constraint on the distribution of zeroes.

To take into account finite size distortions, we can introduce a transformation

\[ m(\gamma) = \gamma - \Delta m(\gamma) \tag{17} \]

which is the identity on the real axis and which maps the zeroes \(\gamma_n\) of the partition sum onto a uniform density perpendicular to the real axis

\[ m(\gamma_n) = \lambda_0 + i(2n + 1)\frac{\pi}{N\delta} \tag{18} \]

As before we can introduce an analytic function \(\bar{f}(\gamma)\) such that

\[ Z_\gamma = 2 \cosh \left( (m(\gamma) - \lambda_0) \frac{N\delta}{2} \right) \bar{f}(\gamma) \tag{19} \]

Using this expression in the inverse Fourier transform eq.(12) the probability distribution at the transition point reads

\[ p_{\lambda_0}(b) = \bar{g}^+_{\lambda_0}(b + \frac{N\delta}{2}) + \bar{g}^-_{\lambda_0}(b - \frac{N\delta}{2}) \tag{20} \]

where

\[ \bar{g}^\pm_{\lambda_0}(b) = N_0 \int d\eta \bar{f}(\lambda_0 + i\eta)e^{\mp \Delta m(\lambda_0+i\eta)N\delta/2}e^{i\eta b} \tag{21} \]

If a phase transition is present at the thermodynamic limit, according to the Yang-Lee theorem \(\Delta m\) goes to zero faster than \(N\) such that the two \(\bar{g}^\pm_{\lambda_0}\) converge to the same distribution \(g_{\lambda_0}\), and \(p_{\lambda_0}(b)\) tends to eq.(13). Following the discussion of eq.(20), this means that a deformed distribution of zeroes that converges to a straight line of equally spaced zeroes perpendicular to the real axis gives rise to a bimodal probability distribution function.
In finite systems, if $\Delta m$ is not zero, the sum (20) may or may not present a concavity anomaly depending on the actual properties of $\Delta m$. This is true irrespectively of the fact that the straight distribution of zeroes eq.(10) is asymptotically reached (i.e. a first order phase transition exists in the bulk) or not. In this case, the physical phenomenon could be classified differently with the zeroes of the partition sum [2] or with the concavity criteria [4, 5]. Indeed, we have demonstrated their equivalence only for large systems.

In conclusion, our topological definition of phase transitions through the bimodality of the probability distribution function, generalizes the definition based on the microcanonical entropy [3, 4] and is equivalent to the definition based on convexity anomalies of generic thermodynamic potentials [5] for any number of particles. We have demonstrated that a bimodality is equivalent to the Yang Lee definition of phase transitions close to and at the thermodynamical limit. A uniform distribution of zeroes perpendicular to the real axis of a Lagrange multiplier $\lambda$ at the position $\lambda_0$ implies that the associated distribution of events is bimodal for this $\lambda_0$, and conversely a bimodal distribution at a given $\lambda_0$ generates a uniform distribution of zeroes with a real part $\lambda_0$ and a regularly spaced imaginary part. This means that the occurrence of bimodalities can be considered as a valid generalization of the concept of phase transition to systems of any size. This topological definition is extremely powerful since it gives access to the order parameters defined as the observables for which the distribution is bimodal. We have demonstrated that this definition agrees with the definition based on the zeroes of $Z$ [2] when the bimodal distribution can be approximated by two gaussians, i.e. in the double saddle point approximation. This means that the different definitions are coherent for $N$ big enough that fluctuations around the two phases are at the gaussian level, but they can differ for smaller systems. The compatibility of the different definitions at the thermodynamic limit means that the great effort done by the different research groups to recognize and classify phase transitions in finite systems [2, 4, 16] is not only rigorous but also consistent with macroscopic thermodynamics.

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