Lyapunov exponents as a dynamical indicator of a phase transition

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Abstract. – We study analytically the behavior of the largest Lyapunov exponent \( \lambda_1 \) for a one-dimensional chain of coupled nonlinear oscillators, by combining the transfer integral method and a Riemannian geometry approach. We apply the results to a simple model, proposed for the DNA denaturation, which emphasizes a first order-like or second order phase transition depending on the ratio of two length scales: this is an excellent model to characterize \( \lambda_1 \) as a dynamical indicator close to a phase transition.

Introduction. – Phase transition plays a central role in equilibrium and non equilibrium statistical physics books and lectures in particular because it exemplifies the paradigmatic concept of universality in physics. In the theory of dynamical systems, the concept of Lyapunov exponent has also attracted a lot of attention because it defines unambiguously a sufficient condition for chaotic instability, but unfortunately except for very few systems it is already an extremely difficult task to derive analytically the expression of the largest one, \( \lambda_1 \), as a function of the energy density. As some promising results have recently been obtained to describe some properties of high-dimensional dynamical systems, by combining tools developed in the framework of dynamical systems with concepts and methods of equilibrium statistical mechanics, the idea that both concepts could be related was proposed recently.

During the last years, the process by which two strands of DNA unbind upon heating, called DNA denaturation or melting, has motivated a lot of works and in particular an extremely simplified dynamical model was proposed and studied. This one-dimensional Hamiltonian has the following expression

\[
H = \sum_n \frac{m}{2} \dot{y}_n^2 + \frac{K}{2} (y_n - y_{n+1})^2 + D (e^{-a y_n} - 1)^2
\]

(1)

where \( m \) corresponds to the effective mass of nucleotides, \( K \) the coupling constant and \( D \) (resp. \( a \)) the depth (resp. inverse length scale) of the Morse potential which mimic the interactions between groups of atoms of opposite strands.

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This potential with only nearest-neighbor interactions is well suited for using the transfer integral method in order to derive the canonical partition function. It can be shown [12] that this system can be mapped to the quantum mechanical analogy of a particle in a Morse potential and that all thermodynamical quantities could finally be expressed as functions of the eigenvalues $\varepsilon_q$ and eigenfunctions $\phi_q$ of this Schrödinger problem. In particular, we showed the existence of a critical temperature $T_c = 2\sqrt{2KD/ak_B}$ corresponding to a second order phase transition between the so-called native (or double strand) state with the particles in the bottom of the Morse well, and the denaturated state, with the particles on the Morse plateau.

The dynamical properties of this one-dimensional model are also very interesting and have, in particular, emphasized the role of localized oscillating excitations, called discrete breathers, as precursors effects driving this phase transition. Spatiotemporal studies of the dynamics reveals also intermittency-like features and has led us to consider the importance of its chaotic properties as an important ingredient to characterize and to explain this dynamical instability. This is the reason why it is important to study the Lyapunov behavior as a function of the energy density, not only by computing it numerically but also (if possible!) by deriving its expression using the Riemannian geometry approach proposed recently [6].

Riemannian Geometry Approach. – The main idea is that the chaotic hypothesis is at the origin of the validity of equilibrium statistical physics, and this should be traced somehow in the dynamics and therefore in the largest Lyapunov exponent. Through a reformulation of Hamiltonian dynamics in the language of Riemannian geometry [6], the method proposes therefore to relate the microscopic dynamics to the statistical averages [13]. Applied to a typical high-dimensional hamiltonian system, this corresponds to study the parametric-like instability of geodesics, driven by the fluctuations of the curvature. Indeed, chaos can be induced not only by negative curvatures but also by positive ones, provided they are fluctuating, as it is less well known. Once the curvature $\kappa_0$ and its fluctuations $\sigma_\kappa$ are accurately determined, the method uses a gaussian stochastic process to approximate the effective curvature felt along a geodesic and finally one ends up with the following expression of the largest Lyapunov exponent

$$\lambda_1 = \frac{1}{2} \left( \Lambda - \frac{4\kappa_0}{3\Lambda} \right) \text{ where } \Lambda = \left( \frac{\sigma_\kappa^2 \tau}{3} + \sqrt{\left( \frac{4\kappa_0}{3} \right)^3 + \sigma_\kappa^4 \tau^2} \right)^{1/3}. \quad (2)$$

In this definition, $\tau$, the relevant time scale associated to the stochastic process is function of the two following timescales: $\tau_1 \simeq \pi/2\sqrt{\kappa_0 + \sigma_\kappa}$ is the time needed to cover the distance between two successive conjugate points along the geodesics, whereas $\tau_2 \simeq \sqrt{\kappa_0}/\sigma_\kappa$ is related to the local curvature fluctuations. The general rough physical estimate $\tau \simeq (1/\tau_1 + 1/\tau_2)^{-1}$ completes finally the analytical estimate of $\lambda_1$ that we would like to continue now by the calculation of the mean value of the curvature and its fluctuations as a function of the energy density.

Curvature and fluctuations along geodesics. – Using the simplifying Eisenhart metric, the Ricci curvature $K_R(y)$ corresponds to the Laplacian of the potential energy. Defining the function $g(y) = 2e^{-2ay} - e^{-ay}$, we get the following expression

$$\kappa_0 = \frac{(K_R)_\mu}{N-1} \simeq 2K + 2a^2 D(g(y))_\mu = 2K + 2a^2 D \left( 1 - \frac{T}{T_c} \right) \quad \text{if } T < T_c, \quad (3)$$

$$= 2K \quad \text{if } T > T_c \quad (4)$$
Fig. 1 – Evolution of $\kappa_0$ (circles) and $\sigma_\kappa$ (squares) as a function of the temperature $T$. Circles and squares correspond to microcanonical results, the solid lines to results obtained with the transfer integral method; the dashed line to the analytical expression (3). Finally, the dotted line correspond to the low temperature approximation (12) of the fluctuations of curvature.

where we have used the equality of mean values in microcanonical $\langle \bullet \rangle_\mu$ and canonical $\langle \bullet \rangle_{can}$ ensembles [15]. The last expression is in particular derived using the expression of the ground-state $\phi_0(y)$ of the quantum mechanical analogy since $\langle g(y) \rangle_{can} = \langle \phi_0 | g(y) | \phi_0 \rangle_{can}$. Figure 1 attests that the agreement is excellent between the above analytical expression, the canonical transfer integral results and microcanonical molecular dynamics simulations, obtained using the best 4th order symplectic integrator due to MacLahan-Attela [14].

It is interesting to note that, the mean curvature is always positive whereas the local one is negative close to the inflexion point of the Morse potential. In addition, expression (3) emphasizes the role of discreteness since the curvature would be almost constant in the continuum limit $a^2 D \ll K$. This will have further consequences on the evolution of $\lambda_1$ when the discreteness is changed.

Fluctuations of curvature. – Since numerical computations are simpler in the microcanonical ensemble, while analytical calculations are simpler in the canonical one, to get the fluctuations of the curvature in the microcanonical ensemble, one needs to add the following corrective term [13]

$$\langle \delta^2 K_R \rangle_\mu = \langle \delta^2 K_R \rangle_{can} - \left( \frac{\partial U}{\partial \beta} \right)^{-1} \left( \frac{\partial (K_R)_{can}}{\partial \beta} \right)^2 . \quad (5)$$

The expression of the energy

$$\frac{U}{N} = \frac{1}{2\beta} - \frac{1}{N} \frac{\partial \ln Z_{can}}{\partial \beta} = k_B T + D \frac{T^2}{T_c^2} \quad \text{if} \quad T < T_c, \quad (6)$$

$$= k_B T \quad \text{if} \quad T > T_c, \quad (7)$$
emphasizes the second order of the phase transition, and finally using the expression of the mean value of the curvature, we obtain the microcanonical/canonical correction

$$\langle \delta^2 K_R \rangle_{\mu} - \langle \delta^2 K_R \rangle_{can} = -\frac{4a^4D^2N}{1 + \sqrt{\frac{\omega^2D}{2K T_c}}} \frac{T^2}{T_c^2}$$  \hspace{1cm} (8)

valid below $T_c$. This quadratic correction vanishes for very low temperature and converges toward a negative constant close to $T_c$.

The expression of the fluctuations of curvature in the canonical ensemble is then

$$\langle \delta^2 K_R \rangle_{can} = \langle (K_R^2(y) - \langle K_R(y) \rangle_{can})^2 \rangle_{can} = 4a^4D^2 \sum_{i,j} \left( \langle g(y_i)g(y_j) \rangle_{can} - \langle g(y_i) \rangle_{can} \langle g(y_j) \rangle_{can} \right).$$

The second part of the parenthesis could be easily computed and gives a term $N^2 \langle g \rangle^2$. The first one can be evaluated with the transfer integral method since, thanks to the periodic boundary condition, one needs to take into account only the difference in indices ($i - j$). However, one cannot integrate directly the integral over the resulting $(N-1)$ variables, and one needs to decompose twice on the orthonormal basis $\{ \phi_q \}$ of the transfer operator. Defining the matrix elements of $g(y)$ in this basis

$$M_{k,q} = \int_{-\infty}^{+\infty} \phi_{q_0}^* (y) g(y) \phi_k(y) dy = \langle \phi_{q_0} | g(y) | \phi_k \rangle, \hspace{1cm} (9)$$

we finally obtain

$$\langle g(y_p)g(y_N) \rangle_{can} = \frac{1}{Z_e} \sum_{k,q} e^{-(N-p)\beta(\varepsilon_q - \varepsilon_k)} e^{-N\beta \varepsilon_k} |M_{k,q}|^2.$$

As only the lowest eigenvalue $\varepsilon_0$ will contribute in the thermodynamic limit, we finally get

$$\langle \delta^2 K_R \rangle_{can} = 4a^4D^2N \sum_{q=1}^{+\infty} \frac{|M_{0,q}|^2}{1 - e^{-\beta(\varepsilon_q - \varepsilon_0)}}$$ \hspace{1cm} (10)

by recognizing a geometrical sum. Figure (1) emphasizes the excellent agreement between microcanonical simulations (squares) and the above analytical expression (solid line) where we have used the eigenfunctions of the transfer integral operator.

Low temperature approximation. – The above expression (10) is however difficult to handle in general but could be simplified in the low temperature regime since the gap between the eigenvalue of the ground state $\varepsilon_0$ and the other ones justify to neglect the exponential term in the denominator. This approximation breaks down of course close to the phase transition since the critical temperature is defined by the disappearance of the ground state $\varepsilon_0$ in the continuum. We obtain thus

$$\langle \delta^2 K_R \rangle_{can} \simeq 4a^4D^2N \sum_{q=1}^{+\infty} |\langle \phi_0 | g | \phi_q \rangle|^2 = 4a^4D^2N \left[ \langle g^2 \rangle - \langle g \rangle^2 \right].$$ \hspace{1cm} (11)

Using the procedure described for the calculation of $\langle g \rangle$, one obtains the formula $\langle g^2 \rangle = \left( 1 - \frac{T}{T_c} \right) \left( 1 + \frac{T}{T_c} + \frac{T^2}{T_c^2} \right)$, where the prefactors 2 and 4 have been renormalized to be in
agreement with numerical simulations; the discreteness of the chain, which drives the phase transition, is the main reason for this renormalization. Adding finally the microcanonical correction \( \sigma_k^2 \equiv \langle \delta^2 K \rangle_{\mu} = 4a^4D^2 \left[ \left( 1 - \frac{T}{T_c} \right) \left( \frac{5T}{T_c} + \frac{2T^2}{T_c^2} \right) - \frac{1}{1 + \sqrt{\frac{a^2D}{2K} \frac{T}{T_c}}} \right] \). \( (12) \)

The dotted line plotted in Fig. (1) attests the quality of this low temperature approximation which leads to a linear dependence versus temperature of the fluctuations \( \sigma_k^2 \sim 20a^4D^2T/T_c \) at the lowest order.

In the neighborhood of the phase transition, the last bound state is too close to the continuum: the exponential term in the denominator of Eq. (10) cannot be neglected and one needs to take into account fully the interactions with the continuum. Due to the rapid oscillations of the scattering eigenstates, one could use the approximate expression for the phase shift of the scattering states, as it was successfully shown in the calculation of the static structure factor and therefore for the susceptibility as its limit \( [12] \). But, unfortunately for the present calculation, there is an important contribution of eigenstates with nonvanishing eigenvectors which prevents this high temperature approximation to succeed.

Largest Lyapunov exponent. – Once the curvature \( (3) \) and its fluctuations \( (10) \) have been calculated, it is straightforward to compute the two timescale \( \tau_1 \) and \( \tau_2 \). Having in mind that the estimate of the decorrelation time scale \( \tau \) is still somehow rudimentary \( [6] \) in the Riemannian framework outlined above, Fig. (4) shows that the alternative definition \( \tau \simeq \left( \frac{1}{\tau_1} + \frac{5}{\tau_2} \right)^{-1} \) gives a particularly striking agreement between the microcanonical numerical results obtained using the standard algorithm \( [16] \) and the analytical derivation proposed here. Let us emphasize that the agreement is excellent on the whole interval contrary to earlier results obtained for the \( \phi^4 \) chain \( [17] \). The approach described here has a domain of validity even larger than expected since the agreement is very good even in the region where the fluctuations of curvature are of the order or greater than the curvature itself.

It is also very interesting to remark that the simultaneous use of the Riemannian geometry approach and the transfer integral method, that we propose here, could be easily extended to the treatment of any chain of oscillators with on-site potential and nearest-neighbor harmonic coupling. Once the transfer integral operator is solved analytically as it is possible here for Hamiltonian \( (1) \) or numerically in general, the evolution of the largest maximal Lyapunov exponent as a function of the energy density is directly derived from expressions of the curvature \( (3) \) and fluctuations \( (10) \), by simply replacing \( 2a^2Dg(y) \) by the second derivative of the onsite potential.

A closer look at very low energy density, shows however a non surprising disagreement since the inset plotted in Fig. (2) clearly violates the low energy density approximate law \( \lambda_1 \sim \sigma_k^2 \), which would give here a linearly increasing function of the temperature according to the lowest order estimate of \( \sigma_k \). The reason is presumably the breakdown of the stochastic approximation for the average curvature in this very low energy limit where the system is almost quadratic and therefore the crucial ergodic hypothesis is not completely fulfilled any more. More interestingly, the numerical model emphasizes an unusual \( 3/2 \) power law contrary to the generally reported \( u^2 \) dependence of \( \lambda_1 \) in high-dimensional dynamical systems \( [6, 7] \).

Let us note that the expression of \( \kappa_0 \) is strongly dependent of the discreteness parameter \( a^2D/K \), but the fluctuations \( \sigma_k \) are not. As Fig. (2) shows that, away from the critical region, smaller the curvature is, greater the Lyapunov is, one can expect that the more discrete the
Fig. 2 – Evolution of the maximal Lyapunov exponent as a function of the energy density $u = U/N$. The symbols correspond to microcanonical numerical simulations for $N = 500$ (triangles), $N = 10^3$ (squares), $N = 2.10^3$ (crosses) and $N = 10^4$ (diamonds). The solid line corresponds to the analytical estimation using the transfer integral method whereas the dashed line corresponds to the analytical expression valid at low energy. The vertical dotted line shows the position of the critical energy density $u_c = k_B T_c + D$. In the inset, we plot $\lambda_1$ as a function of the energy density $u$ in log-log scale. The symbols correspond to microcanonical numerical simulations for $N = 500$ whereas the solid line to a $3/2$ power law.

Fig. 3 – Lyapunov exponent and order of the phase transition. The solid line shows the evolution of the harmonic model with second order phase transition, whereas the symbols are referring to the model with anharmonic coupling with a first-order like phase transition. The number of sites in the chain is $N = 500$ (crosses), $N = 10^3$ (diamonds) and $N = 10^4$ (circles). The vertical dotted (resp. dash-dotted) line corresponds to the critical energy density of Hamiltonian with harmonic (resp. anharmonic) coupling.

chain is, the more chaotic its dynamics would be. This result is clearly in qualitative agreement with the discovery that spontaneously created discrete breathers are actually chaotic excitations and that their domain of stability is far from the continuum limit [18].

Lyapunov and Phase transition. – This model is also a very interesting case where one can characterize $\lambda_1$ as a dynamical indicator of a phase transition. In this case of a second order phase transition, one obtains a critical slowing down reminiscent of the results obtained for the Heisenberg Mean-field model derived by Firpo [6]. We would like also to emphasize the singular behavior of $\kappa_0$, $\sigma_\kappa$ and $\lambda_1$ at the critical energy density, in complete agreement with the exciting conjectures [7] linking them to a topology change in the underlying manifold, being itself an indicator of a thermodynamic phase transition.

More interestingly, we also have studied Hamiltonian \( \mathcal{H} \) with an anharmonic coupling potential \( \frac{k}{2} \left[ 1 + e^{-\alpha(y_n + y_{n-1})} \right] (y_n - y_{n-1})^2 \) which has the property to describe the varying backbone stiffness of the DNA. This hamiltonian was shown to have a first order like phase transition [19] when the ratio of the two inverse length scales $\alpha/a$ is lower than $1/2$. If analytical estimate of the Lyapunov exponent are not possible, Fig. 3 shows its evolution obtained using microcanonical molecular dynamics simulations. In the low energy limit, the
additional parenthesis do not produce of course modifications, but it emphasizes an abrupt change [20] of the Lyapunov exponent at the critical energy density as if $\lambda_1$ was a dynamical order parameter indicating the first order phase transition.

Conclusion. – In this letter, we have presented one of the very few analytical calculation of the largest Lyapunov exponent in a high-dimensional dynamical system [2]. As expected, in addition to serve as a criterion for chaos, or as a characteristic time scale of chaoticity, $\lambda_1$ is an excellent dynamical indicator of the presence of the phase transition and could be used not only to describe the dynamics but also the statistical properties of high dimensional systems.

It is also interesting to consider the behavior of another important dynamical indicator, the participation ratio of the normalized Lyapunov vector $V_1$ associated to the maximal Lyapunov $\lambda_1$. Defined as $\xi = 1/\left(N \sum_{i=1}^{N} [V_1(i)^2 + V_1(i + N)^2]^{\frac{1}{2}}\right)$, where the first (resp. last) $N$ components are associated to the evolution of linear perturbations of $y_n$ (resp. $\dot{y}_n$) in tangent space, $\xi$ is an indicator of localization [21, 22]: it is of order one if the vector is extended and of order $1/N$ if localized. Here, we found that the phase transition corresponds to a crossover from a localized state in tangent space at low energy density to a more extended state just after the phase transition, confirming that the tangent space trough the largest Lyapunov exponent $\lambda_1$, but also its associated eigenvector $V_1$ through the participation ratio $\xi$, are surprisingly good dynamical indicators for emphasizing thermodynamical phase transitions.

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