

Lyapunov exponents as a dynamical indicator of a phase transition

J. BARRÉ and T. DAUXOIS(*)

*Laboratoire de Physique, UMR CNRS 5672, Ecole Normale Supérieure de Lyon
46, allée d'Italie, 69364 Lyon cédex 07, France*

(received 19 February 2001; accepted in final form 3 May 2001)

PACS. 05.45.-a – Nonlinear dynamics and nonlinear dynamical systems.

PACS. 05.70.Fh – Phase transitions: general studies.

PACS. 02.40.-k – Geometry, differential geometry, and topology.

Abstract. – We study analytically the behavior of the largest Lyapunov exponent λ_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 λ_1 as a dynamical indicator close to a phase transition.

Introduction. – *Phase transition* plays a central role in equilibrium and nonequilibrium 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 [1,2] 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, λ_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 [3–6], 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 [7].

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 [8–11] and in particular an extremely simplified dynamical model [12] was proposed and studied. This one-dimensional Hamiltonian has the following expression:

$$H = \sum_n \frac{m}{2} \dot{y}_n^2 + \frac{K}{2} (y_n - y_{n+1})^2 + D (e^{-ay_n} - 1)^2, \quad (1)$$

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

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 has been shown [12]

(*) E-mail: Thierry.Dauxois@ens-lyon.fr

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 ε_q and eigenfunctions ϕ_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 reveal also intermittency-like features and have 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 [7].

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. The method is based on a reformulation of Hamiltonian dynamics in the language of Riemannian geometry [7]: the trajectories are seen as geodesics of a suitable Riemannian manifold. The chaotic properties of the dynamics are then directly related to the curvature of the manifold and its fluctuations. Indeed, negative curvatures tend to separate initially close geodesics, and thus imply a positive Lyapunov exponent; nevertheless chaos may also be induced by positive curvatures, provided they are fluctuating, through a parametric-like instability. To approximate the curvature felt along a geodesic, the method uses a Gaussian statistical process. The mean value of this process is given by κ_0 and its variance by σ_κ , where κ_0 and σ_κ are the statistical average of the curvature and its fluctuations, which can be computed by standard methods of statistical mechanics. Using a result in stochastic differential equations, one finally ends up with the expression of the largest Lyapunov exponent

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

In this definition, τ , the relevant time scale associated to the stochastic process, is a function of the two following time scales: $\tau_1 \simeq \pi/2\sqrt{\kappa_0 + \sigma_\kappa}$ 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 λ_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 $\mathbf{K}_R(\mathbf{y})$ corresponds to the Laplacian of the potential energy. Defining the function $g(y) = 2e^{-2ay} - e^{-ay}$, we get the following expression:

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

$$= 2K, \quad \text{if } T > T_c, \quad (4)$$

where we have used the equality of mean values in microcanonical $\langle \bullet \rangle_\mu$ and canonical $\langle \bullet \rangle_{\text{can}}$ ensembles [13]. The last expression is in particular derived using the expression of the ground-state $\phi_0(y)$ of the quantum-mechanical analogy since $\langle g(y) \rangle_{\text{can}} = \langle \phi_0 | g(y) | \phi_0 \rangle_{\text{can}}$. 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 λ_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]:

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

The expression of the energy

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

$$= k_B T, \quad \text{if } T > T_c, \quad (7)$$

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

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

valid below T_c . This quadratic correction vanishes for very low temperatures and converges towards a negative constant close to T_c .

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

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

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 base

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

we finally obtain

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

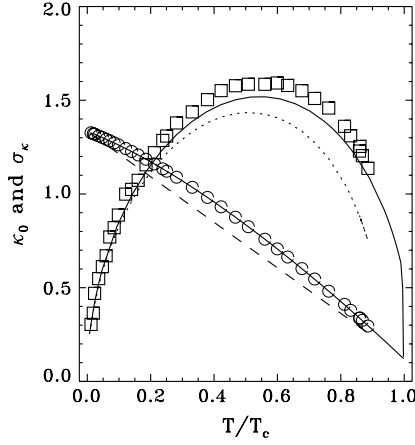


Fig. 1 – Evolution of κ_0 (circles) and σ_κ (squares) as a function of the temperature T . Circles and squares correspond to microcanonical results, the solid lines to results obtained with the transfer integral method (eq. (10)); the dashed line to the analytical expression (eq. (3)). Finally, the dotted line corresponds to the low-temperature approximation (eq. (12)) of the fluctuations of the curvature. Even if these quantities are microcanonical ones, for clarity purpose, we plot them *vs.* T which appears more naturally in the calculations.

As only the lowest eigenvalue ε_0 will contribute in the thermodynamic limit, we finally get

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

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 ε_0 and the other ones justify neglecting 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 ε_0 in the continuum. We thus obtain

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

Using the procedure described for the calculation of $\langle g \rangle$, one obtains the formula $\langle g^2 \rangle = \left(1 - \frac{T}{T_c}\right) \left(1 + \frac{4T}{T_c} + \frac{2T^2}{T_c^2}\right)$, 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 (8), we obtain

$$\sigma_\kappa^2 \equiv \frac{\langle \delta^2 K_R \rangle_\mu}{N} = 4a^4 D^2 \left[\left(1 - \frac{T}{T_c}\right) \left(\frac{5T}{T_c} + \frac{2T^2}{T_c^2}\right) - \frac{1}{1 + \sqrt{\frac{a^2 D}{2K} \frac{T}{T_c}}} \frac{T^2}{T_c^2} \right]. \quad (12)$$

The dotted line plotted in fig. 1 attests the quality of this low-temperature approximation which leads to a linear dependence *vs.* temperature of the fluctuations $\sigma_\kappa^2 \sim 20a^4 D^2 T/T_c$ at the lowest order.

Largest Lyapunov exponent. – Once the curvature (3) and its fluctuations (10) have been calculated, it is straightforward to compute the two time scales τ_1 and τ_2 . Having in mind that the estimate of the decorrelation time scale τ is still somehow rudimentary [7] in the Riemannian framework outlined above, fig. 2 shows that the alternative definition $\tau \simeq (1/\tau_1 + 5/\tau_2)^{-1}$ gives a particularly striking agreement between the microcanonical numerical results obtained using the standard algorithm [15] 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 ϕ^4 chain [16]. 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 of or greater than the curvature itself.

It is also very interesting to remark that the simultaneous use of the Riemannian geometry approach and of 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 $2a^2 Dg(y)$ by the second derivative of the on-site potential.

A closer look at very low-energy density shows, however, a nonsurprising disagreement since the inset plotted in fig. 2 clearly violates the low-energy density approximate law $\lambda_1 \sim \sigma_\kappa^2$, which would give here a linearly increasing function of the temperature according to the lowest-order estimate of σ_κ . 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 λ_1 in high-dimensional dynamical systems [6, 7].

Let us note that the expression of κ_0 is strongly dependent on the discreteness parameter $a^2 D/K$, but the fluctuations σ_κ are not. As fig. 2 shows that, away from the critical region, the smaller the curvature is, the greater the Lyapunov is, one can expect that the more discrete the 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 [17].

Lyapunov and phase transition. – This model is also a very interesting case where one can characterize λ_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 κ_0 , σ_κ and λ_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 the Hamiltonian (1) with an anharmonic coupling potential $\frac{K}{2}[1 + e^{-\alpha(y_n + y_{n-1})}](y_n - y_{n-1})^2$ 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 [18] when the ratio of the two inverse length scales α/a is lower than $1/2$. If analytical estimates of the Lyapunov exponent are not possible, fig. 3 shows its evolution obtained using microcanonical molecular dynamics simulations. In the low-energy limit, the

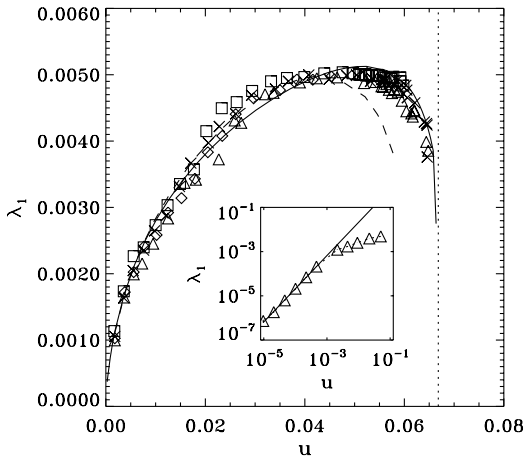


Fig. 2

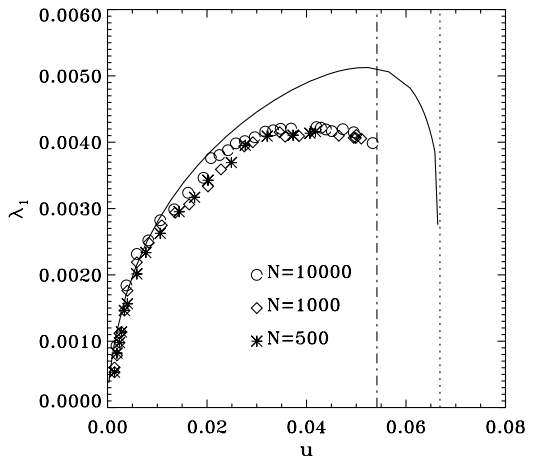


Fig. 3

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), $N = 2 \cdot 10^3$ (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 λ_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 (respectively, dash-dotted) line corresponds to the critical energy density of Hamiltonian with harmonic (respectively, anharmonic) coupling.

additional parenthesis does not produce, of course, modifications, but it emphasizes an abrupt change [19] of the Lyapunov exponent at the critical energy density as if λ_1 were 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, λ_1 is an excellent dynamical indicator of the presence of the phase transition and could be used to describe not only 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 λ_1 . Defined as $\xi = 1 / \left(N \sum_{i=1}^N [V_1(i)^2 + V_1(i+N)^2]^2 \right)$, where the first (respectively, last) N components are associated to the evolution of linear perturbations of y_n (respectively, \dot{y}_n) in tangent space, ξ is an indicator of localization [20–22]: 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 through the largest Lyapunov

exponent λ_1 , but also its associated eigenvector V_1 through the participation ratio ξ , are surprisingly good dynamical indicators for emphasizing thermodynamical phase transitions.

* * *

We wish to acknowledge very helpful discussions with A. ALASTUEY, J. FARAGO, M.-C. FIRPO, S. LEPRI, R. LIVI, M. PEYRARD, S. RUFFO, M. TERRANEO, N. THEODORAKOPOULOS. This work has been partially supported by EU Contract No. HPRN-CT-2000-00163 (LOCNET network).

REFERENCES

- [1] MANNEVILLE P., *Structures Dissipatives, Chaos et Turbulence* (Aléa Saclay) 1991.
- [2] LICHTENBERG A. J. and LIEBERMAN M. A., *Regular and Chaotic Dynamics* (Springer, Berlin) 1992.
- [3] CASETTI L., LIVI R. and PETTINI M., *Phys. Rev. Lett.*, **74** (1995) 375.
- [4] CONSTANTOUDIS V. and THEODORAKOPOULOS N., *Phys. Rev. E*, **55** (1997) 7612.
- [5] LATORA V., RAPISARDA A. and RUFFO S., *Physica D*, **131** (1999) 38.
- [6] FIRPO M. C., *Phys. Rev. E*, **57** (1998) 6599.
- [7] CASETTI L., PETTINI M. and COHEN E. G. D., *Phys. Rep.*, **337** (2000) 237.
- [8] COCCO S. and MONASSON R., *Phys. Rev. Lett.*, **83** (1999) 5178.
- [9] CAUSO M. S., COLUZZI B. and GRASSBERGER P., *Phys. Rev. E*, **62** (2000) 3958.
- [10] KAFRI Y., MUKAMEL D. and PELLITI L., *Phys. Rev. Lett.*, **85** (2000) 4988.
- [11] GAREL T., MONTHUS C. and ORLAND H., cond-mat/0101058.
- [12] THEODORAKOPOULOS N., DAUXOIS T. and PEYRARD M., *Phys. Rev. Lett.*, **85** (2000) 6.
- [13] LEBOWITZ J. L., PERCUS J. K. and VERLET L., *Phys. Rev.*, **153** (1967) 250.
- [14] The parameters of the model are $D = 0.03$ eV, $K = 0.06$ eV/Å², $a = 4.5$ Å⁻¹, $\alpha = 0.35$ Å⁻¹ and $m = 300$ a.m.u.
- [15] BENETTIN G. *et al.*, *Meccanica*, **9** (1980) 21.
- [16] CAIANI L. *et al.*, *Phys. Rev. E*, **57** (1998) 3886. 7
- [17] MAC KAY R. S. and AUBRY S., *Nonlinearity*, **7** (1994) 1623.
- [18] Results reported in [12] clearly shows the behavior of a first-order phase transition with an unusual diverging correlation length. However, it is likely that the order of the transition would be second order in the critical region which is extremely thin and unreachable numerically. See ZIA R. K. P., LIPOWSKY R. and KROLL D. M., *Am. J. Phys.*, **56** (1988) 160 or THEODORAKOPOULOS N., unpublished (2000). However, as the width of this critical region could be decreased only by decreasing the ratio α/a , this phase transition will be considered as first-order-like.
- [19] MEHRA V. and RAMASWAMY R., *Phys. Rev. E*, **56** (1997) 2508.
- [20] The strong stochasticity threshold of the Fermi-Pasta-Ulam chain, characteristic of the transition between low- and strong-chaotic regime, was shown to be closely related to a transition in tangent space, the Lyapunov eigenvector V_1 being more localized in space at high energy. See DAUXOIS T., RUFFO S. and A. TORCINI, *Phys. Rev. E*, **56** (1997) R6229.
- [21] PIKOVSKY A. and POLITI A., *Nonlinearity*, **11** (1999) 1049.
- [22] KOCKELKOREN J. and CHATÉ H., unpublished (2000).