When It Helps to Be Purely Hamiltonian: Acceleration of Rare Events and Enhanced Escape Dynamics

Dirk Hennig[1], Simon Fugmann[1], Lutz Schimansky-Geier[1], and Peter Hänggi[2]

[1] Institut für Physik, Humboldt-Universität zu Berlin, Newtonstr. 15, 12489 Berlin, Germany
hennigd@physik.hu-berlin.de

[2] Institut für Physik, Universität Augsburg, Universitätsstraße 1, 86135 Augsburg, Germany
hanggi@physik.uni-augsburg.de

Abstract. We consider the self-organized escape of a linear chain of coupled units from a metastable state over a barrier in a microcanonical situation. Initially the units of the chain are situated near the bottom of the potential well forming a flat state. In the underlying conservative and deterministic dynamics such a uniform and linear lattice state with comparatively little energy content seems to be restrained to oscillations around the potential bottom preventing escape from the well. It is demonstrated that even small deviations from the flat state entail internal energy redistribution leading to such strong localization that the lattice chain spontaneously adopts a localized pattern resembling a hairpin-like structure. The latter corresponds to a critical equilibrium configuration, that is a transition state, and, being dynamically unstable, constitutes the starting point for the escape process. The collective barrier crossing of the units takes place as kink-antikink motions along the chain. It turns out that this nonlinear barrier crossing in a microcanonical situation is more efficient compared with a thermally activated chain for small ratios between the total energy of the chain and the barrier energy.

1 Introduction

The intensively investigated Kramers problem concerns the escape of a Brownian particle from a metastable state over a barrier (for a review see [1]). There it is implied that the system is in contact with an external heat bath serving as a permanent energy source, causing dissipation and local energy fluctuations which can trigger the escape process. However, the occurrence of the necessary optimal fluctuations enabling the particle to stochastically overcome an energetic barrier can be a rare event. This is particularly the case when the ratio between the thermal energy supplied to the particle by

the heat bath and the barrier energy is low and the rate of thermal barrier crossing is exponentially suppressed. The extension of the escape problem to multi-dimensional systems was given in [2]. Recently, in the biophysical context there has been growing interest in thermally activated barrier crossing of a polymer chain occurring e.g. during the transport of long and flexible polymers across membranes and DNA electrophoresis [3–6].

Different to the numerous studies of the thermally induced escape, based on the coupling of the system to an external heat bath, the microcanonical situation, when only the internal energy of a system has to suffice to perform structural transitions, has been studied less. The question is whether under deterministic and conservative conditions a coupled oscillator chain can still overcome a potential barrier when all its units reside initially near the bottom of a metastable potential well? In such a situation the energy is almost equally shared among the units and the system is also far away from the critical equilibrium configuration related with a saddle point in configuration space, referred to as the transition state [7]. Typically, the critical equilibrium configuration is represented by a localized state of the chain.

Concerning the attainment of localized structures it is by now well established that nonlinear systems exhibit localization features giving rise to the formation of coherent structures that emerge even from an initial almost homogeneous state [8]. The concentration of an originally distributed physical quantity to a few degrees of freedom in confined regions of a spatially extended and homogeneous systems proceeds often in a self-organized manner. In recent years the concept of intrinsic localized modes or discrete breathers as time-periodic and spatially localized solutions of nonlinear lattice systems has turned out to present the prototype of excitations describing localization phenomena in numerous physical situations [9–14]. For the creation of localized structures modulational instability leading to a self-induced modulation of an initial linear wave with a subsequent generation of localized pulses has proven to be an effective mechanism. In this way energy localization in a homogeneous system is achievable. For example breathers have been successfully applied to describe localized excitations which reproduce typical features of the thermally induced opening dynamics of DNA duplex molecules such as the magnitude of the amplitude and the time-scale of the oscillating bubble preceding full strand separation (denaturation) [15–21].

In the present study we address the escape problem of a one-dimensional oscillator chain over the barrier of a metastable potential within a conservative and deterministic lattice model [22, 23]. The units of the lattice system are initially near a metastable equilibrium which hinders the system to immediately perform a task that is associated with overall large-amplitude excitations. In more detail, we consider the energy exchange dynamics in a nonlinear oscillator chain. Each oscillator evolves in a local anharmonic potential possessing a barrier that divides regions of bounded motion in the potential well from unbounded ones beyond the barrier. The oscillators are linearly coupled. Concerning the energy redistribution we focus interest on the initial situation of

a low-energy mode of the chain. The question which arises now is whether with the low amount of energy supplied to the system the focusing of energy proceeds that effectively that at least one, if not a few units, can gain enough energy to get over the potential barrier? If so, is it possible that the neighbors coupled to the escaping unit(s) also get drawn over the barrier? If more and more units get involved this would initiate a coordinated escape of the entire chain.

2 The oscillator chain model

The linear chain is modeled as a one-dimensional lattice system of harmonically coupled nonlinear oscillators with the Hamiltonian

$$H = \sum_{n=1}^{N} \left\{ \frac{p_n^2}{2} + U(q_n) \right\} + \frac{\kappa}{2} \sum_{n=1}^{N} \left[q_{n+1} - q_n \right]^2 . \tag{1}$$

The coordinates q_n quantify the amplitude of an oscillator at site n. p_n denotes the momentum canonically conjugate to the coordinate q_n. Each oscillator evolves in an anharmonic potential given by

$$U(q) = \frac{\omega_0^2}{2} q^2 - \frac{a}{3} q^3 , \tag{2}$$

where $a > 0$. The metastable equilibrium of the potential is situated at $q = 0$ and the maximum is located at $q = \omega_0^2/a$. There is a potential barrier which particles have to overcome in order to escape from the potential well of depth $\Delta E = \omega_0^6/(6a^2)$. The oscillators, also referred to as units, are coupled harmonically with nearest-neighbor interaction strength κ. The equations of motion derived from the Hamiltonian given in Eq. (1) read

$$\frac{d^2 q_n}{dt^2} + \omega_0^2 q_n - a q_n^2 - \kappa \left[q_{n+1} + q_{n-1} - 2q_n \right] = 0 . \tag{3}$$

We impose periodic boundary conditions according to $q_{N+1} = q_1$. Note that nonlinearity is solely contained in the local potential term.

3 Spontaneous energy localization

The ability of nonlinear and discrete systems to exhibit spontaneous localization has been demonstrated recently [24–30]. Such formation of localized excitations can be caused by modulational instability leading to the formation of intrinsically localized modes (breathers). This mechanism initiates an instability of an initial linear wave when small perturbations of non-vanishing wavenumbers are imposed. The instability, giving rise to an

exponential growth of the perturbation, destroys the initial linear wave at a critical wavenumber so that a localized hump gets formed.

Initially all the oscillators are placed close to the bottom of the potential well. The chain state is expressed as a plane wave solution (phonons) $q_n(t) = (1/2)q_0 \exp[i(k\,n - \omega t)] + c.c.$ obeying the dispersion relation $\omega^2 = \omega_0^2 + 4\kappa \sin^2(k/2)$ with wave number $k \in [0, \pi]$.

Small modulational perturbations on the plane wave solution are imposed taking random initial amplitudes and/or momenta uniformly distributed in small intervals $|q_n(0) - q_0| \leq \Delta q$ and $|p_n(0) - p_0| \leq \Delta p$, respectively. Thus the chain is initialized close to an *almost homogeneous state* and yet such desynchronized ($\Delta q \neq 0$) to have small but nonvanishing initial interaction terms initiating energy exchange between the coupled units.

We recall that an uniform lattice state with amplitude q_0 and wave number k remains stable as long as the nonlinear character related with the aq^3 term of the potential $U(q)$ can be neglected. The chain evolves harmonically and localization of energy does not take place. Otherwise, the nonlinear part of the potential makes a modulational instability of waves possible. That is perturbations with a wave number Q may grow exponentially resulting in accumulation of energy at the expense of energy from the other units.

The exponential growth for the flat state, i.e. $k = 0$, takes place with rate [22, 23, 28]

$$\Gamma = \sqrt{\sin^2\left(\frac{Q}{2}\right) \frac{\kappa}{\omega_0^2} \left(\frac{5a^2}{3\omega_0^2}\, q_0^2 - 4\kappa\, \sin^2\left(\frac{Q}{2}\right)\right)}\,, \tag{4}$$

if the argument of the square root is positive. Thus it must hold that

$$\frac{5a^2}{3\omega_0^2}\, q_0^2 - 4\kappa\, \sin^2\left(\frac{Q}{2}\right) > 0\,, \tag{5}$$

which means that for fixed q_0 the anharmonicity a needs to be large enough or with given a the q_0 has to obtain overcritical values.

The set of coupled equations (3) has been numerically integrated with a fourth-order Runge-Kutta scheme. The accuracy of the calculation was checked by monitoring the conversation of the total energy with precision of at least 10^{-4}. The chain consists of $N = 100$ coupled oscillators.

Starting from an initial lattice state of nearly equipartition the attainment of a nontrivial structure is observed. More precisely, a pattern evolves in the course of time (of the order of $t \sim 2 \times 10^2$) for which at some lattice sites the amplitudes grow considerably whereas they get lowered in the surrounding regions. This localization phenomenon is reflected in the appearance of an array of irregularly spaced breathers on the lattice as seen in Fig. 1 where the spatio-temporal evolution of the energy density

$$E_n = \frac{p_n^2}{2} + U(q_n) + \frac{\kappa}{4}\left[\,(\,q_{n+1} - q_n\,)^2 + (\,q_{n-1} - q_n\,)^2\,\right]. \tag{6}$$

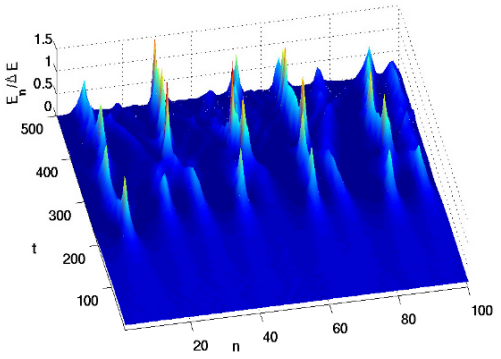

Fig. 1. Spatio-temporal evolution of the energy distribution $E_n(t)$. Initially the coordinates are uniformly distributed in the interval $|q_n(0) - q_0| \leq \Delta q$ with mean $q_0 = 0.45$ and width $\Delta q = 0.01$ and $p_n(0) = \Delta p = 0$. This leads to a total energy $E_{\text{total}} = 17.2 \equiv 12.9 \times \Delta E$. The parameter values are given by $a = 1$, $\omega_0^2 = 2$, $N = 100$ and $\kappa = 0.3$.

is shown. The total energy is $E_{\text{total}} = 17.2$ which is equivalent to $12.9 \times \Delta E$. In the beginning the total energy is virtually evenly shared among all units. The corresponding energy density, i.e. the average amount of energy contained in a single oscillator, lies significantly below the barrier energy. To be precise, the ratio between the energy density and the barrier energy is $E_n(0)/\Delta E = 0.129$. In other words, in order that a unit passes over the energy barrier from the region of bounded into unbounded motion directed flow of energy into this unit is demanded. Actually the energy amount of at least 7 other units has to be transferred into one unit so that this units energy levels that of the barrier.

There exist moving breathers that have the tendency to collide inelastically with other breathers (cf. Fig. 1 at site $n = 30$). In fact, various breathers merge to form larger amplitude breathers proceeding preferably such that the larger amplitude breathers grow on the expense of the smaller ones. As a result energy gets even stronger concentrated into smaller regions of the chain. Such localization scenario has been shown to be characteristic for a number of nonlinear lattice systems [25, 29–33].

For further illustration we depict in Fig. 2 snapshots of the energy density $E_n(t)$ at different instants of time. Starting point is the almost homogeneous state and the first snap shot is taken at $t = 5$ when the pattern is still flat. After a certain time the local energy accumulation is such enhanced that at least one of the involved units possess enough energy to overcome the barrier. As illustrated in Fig. 2 for the snap shot taken at $t = 550$ this happens for the unit at $n = 30$.

The question then is: does an escaped unit continue its flight beyond the barrier or can it even be pulled back into the bound chain formation ($q_n <$

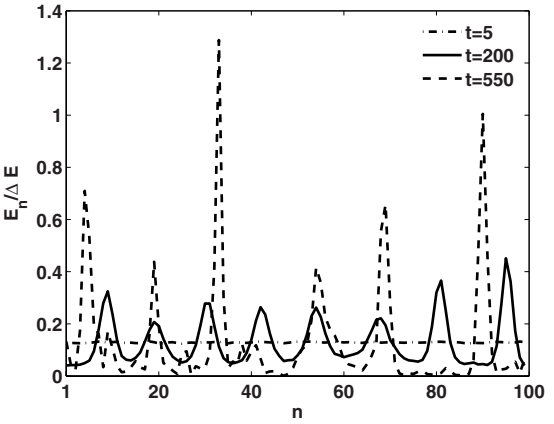

Fig. 2. Snap shots of the energy density at instants of time as indicated in the plot. Initial conditions: $q_0 = 0.45$, $\Delta q = 0.01$, $p_0 = \Delta p = 0$. Parameter values: $\omega_0^2 = 2$, $a = 1$, $N = 100$ and $\kappa = 0.3$.

$q_{\max}$) by the restoring binding forces exercised by its neighbors? On the other hand, the unit that has already escaped from the potential well might drag neighboring ones closer to or in the extreme even over the barrier. Thus, concerted escape of at least parts, if not the whole chain, from the potential valley seems possible.

4 Escape dynamics

Whether a unit of growing amplitude can really escape from the potential well or is held back by the restoring forces of their neighbors depends on the corresponding amplitude ratio as well as on the coupling strength. The critical chain configuration, that is the *transition state*, is determined by $\ddot{q}_n(t) = 0$ resulting in the stationary system

$$-\frac{\partial U}{\partial q_n} + \kappa[q_{n+1} + q_{n-1} - 2q_n] = 0\,. \tag{7}$$

Interpreting n as the 'discrete' time, with $1 \leq n \leq N$, the Eq. (7) describes the motion of a point particle in the inverted potential $-U(q)$. This difference system can be cast in form of a two-dimensional map by defining $x_n = q_n$ and $y_n = q_{n-1}$ [34] which gives:

$$\begin{aligned} x_{n+1} &= (w_o^2 x_n - a x_n^2)/\kappa + 2x_n - y_n \\ y_{n+1} &= x_n\,. \end{aligned} \tag{8}$$

The fixed points of this map are found as $x_0 = y_0 = 0$ and $x_1 = y_1 = \frac{w_0^2}{a}$. A stability analysis reveals that $x_0 = y_0 = 0$ represents an unstable hyperbolic equilibrium while at $x_1 = y_1 = \frac{w_0^2}{a}$ a stable center is located. The map is nonintegrable. The stable and unstable manifold of the hyperbolic point intersect each other yielding homoclinic crossings.

The corresponding homoclinic orbit of the map is on the lattice chain equivalent to a localized hump solution $\tilde{q}_n$ resembling the form of a hairpin. In Fig. 3 the profile of critical equilibrium configurations $\tilde{q}_n$ for several coupling

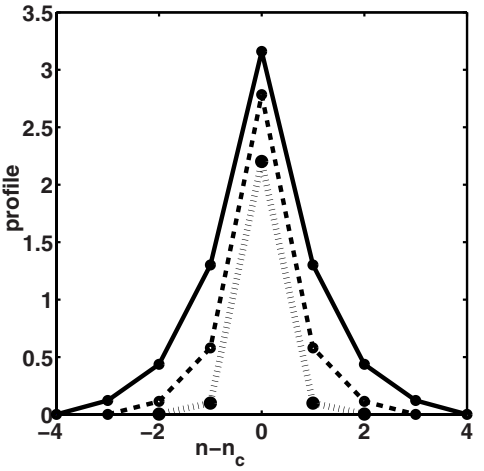

Fig. 3. Amplitude profile of the critical chain configuration for different coupling strengths: $k = 0.1$ (*dashed-dotted line*), $k = 0.5$ (*dashed line*), and $k = 1$ (*solid line*). For better illustration only the part of the lattice chain around the central site n_c with seizable elongations of the bonds is shown.

strengths are displayed. The stronger the coupling is the larger the maximal amplitude of the hump and the wider the width of the latter. Equation (7) can be derived from an energy functional $F = \sum_n \left(U(q_n) + \frac{\kappa}{2}[q_n - q_{n-1}]^2\right)$ as $\partial F/\partial q_n = 0$. Apparently, with increasing coupling more activation energy is needed to get the chain into its critical equilibrium configuration. One obtains $F = 1.33$, $F = 2.77$ and $F = 4.54$ for $\kappa = 0.1$, $\kappa = 0.5$ and $\kappa = 1$, respectively. Most importantly regarding escape, for the elongation of the bond at the central site, i.e. the maximal amplitude $\tilde{q}_{\max}$, it holds that $\partial U(\tilde{q}_{\max})/\partial q < 0$.

In the following we prove the dynamical instability of the critical localized mode. To this end we set $q_n(t) = \tilde{q}_n + w_n(t)$ with $|w_n| \ll 1$ and derive the linearized equations of motion as

$$\ddot{w}_n(t) = -\frac{\partial^2 U(q_n)}{\partial q_n^2}|_{q_n=\tilde{q}_n} w_n(t) + \kappa[w_{n+1}(t) + w_{n-1}(t) - 2w_n(t)]\,. \tag{9}$$

With the ansatz $w_n(t) = \phi_n \exp(\sqrt{\lambda}t)$ for the solution of (9) one arrives at an eigenvalue problem

$$\lambda\phi_n = -V_n\phi_n + \kappa[\phi_{n+1} + \phi_{n-1} - 2\phi_n]\,, \tag{10}$$

with

$$V_n = \frac{\partial^2 U(q_n)}{\partial q_n^2}|_{q_n=\tilde{q}_n} = \omega_0^2 - 2a\tilde{q}_n\,. \tag{11}$$

The second-order difference equation (10) is of the discrete stationary Schrödinger type, with a non-periodic potential, $-V_n$, breaking the translational invariance so that localized solutions exist (so called stop-gap states). The evolution of the two-component vector $(\phi_{n+1}, \phi_n)^T$ is determined by the following Poincaré map:

$$\mathcal{M}:\ \begin{pmatrix}\phi_{n+1}\\ \phi_n\end{pmatrix} = \begin{bmatrix}E_n & -1\\ 1 & 0\end{bmatrix}\begin{pmatrix}\phi_n\\ \phi_{n-1}\end{pmatrix}\,, \tag{12}$$

with on-site energy $E_n = (\lambda + V_n)/\kappa + 2$. The node-less even-parity ground state of Eq. (10), with its energy under the lower edge of the energy band of the passing band states, corresponds to an orbit of the linear map $\mathcal{M}$ being homoclinic to the hyperbolic equilibrium point at the origin $(0, 0)$ of the map plane. For the presence of a hyperbolic equilibrium the following inequality has to be satisfied:

$$\mathrm{Trace}(\mathcal{M}) = E_n = \frac{\lambda + V_n}{\kappa} + 2 > 2\,, \tag{13}$$

implying that λ must fulfill the following constraint:

$$\lambda > \max_n(-V_n) = 2a \max_n \tilde{q}_n - \omega_0^2 > 0. \tag{14}$$

With the maximal amplitude of the c.l.m. lying beyond the barrier, viz. $\max_n \tilde{q}_n > \omega_0^2/a$ one finds

$$\lambda > \omega_0^2 > 0\,. \tag{15}$$

Therefore, the ground state belongs to a positive eigenvalue from which we deduce that perturbations of the corresponding solution in the time domain grow exponentially.

The critical equilibrium solution tell us that for overcritical elongations of the units from their rest positions, $q_n > \tilde{q}_n$, the whole chain performs directed motion over the barrier. Conversely, if the elongations lie below the ones corresponding to $\tilde{q}_n$ then crossing the barrier is excluded.

Since for those states that have passed through the c.l.m. the kinetic energy of the outward motion increases a return backwards over the barrier into the

original well is prevented. Figure 4 illustrates the kink-antikink motion in the escape time of the units $T_{\rm esc}^{(n)}$ (defined as the moment at which a unit passes through a point $q = 20$ far beyond the barrier) versus the position on the lattice. Consecutively all oscillators manages to climb over the barrier one after another in a relatively short time interval.

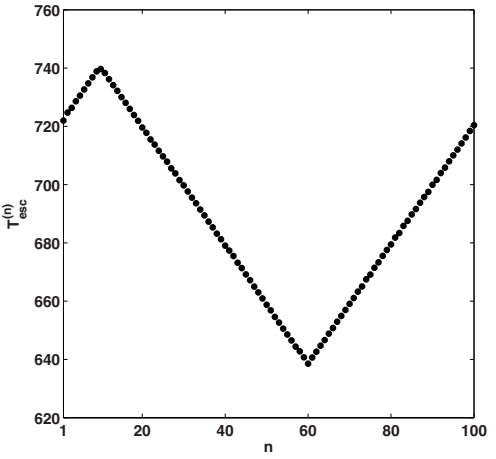

Fig. 4. The escape time of the chain units versus position. Initial conditions: $q_0 = 0.45$, $\Delta q = 0.01$, $p_0 = \Delta p = 0$. Parameter values: $a = 1$, $\omega_0^2 = 2$, $N = 100$ and $\kappa = 0.3$

Finally we compare the microcanonical escape process with a corresponding thermally activated process in the Kramers problem [1]. The Langevin equations read

$$\frac{d^2 q_n}{dt^2} + \gamma \frac{dq_n}{dt} + \frac{dU}{dq_n} - \kappa \left[q_{n+1} + q_{n-1} - 2q_n \right] + \xi_n(t) = 0 \ . \qquad (16)$$

Here γ is the friction parameter and $\xi_n(t)$ is a Gaussian distributed thermal random force with $< \xi_n(t) >= 0$ and $< \xi_n(t)\xi_{n'}(t') >= 2\gamma k_B T \delta_{n,n'} \delta(t - t')$.

Our results are summarized in Fig. 5 showing the mean escape time of the chain. The latter is determined by the mean value of the escape times of its units (see above). We took averages over 500 realizations of random initial conditions in the microcanonical and of noise in the Langevin equations, respectively. The Langevin equations were numerically integrated using a two-order Heun stochastic solver scheme. Results are presented as a function of $E_0/\Delta E$. For the deterministic and conservative system (3) E_0 is given by the initial energy per unit while it corresponds to thermal energy $k_B T$ in case of the Langevin system (16). In both cases there is a rather strong decay of $T_{\rm esc}$

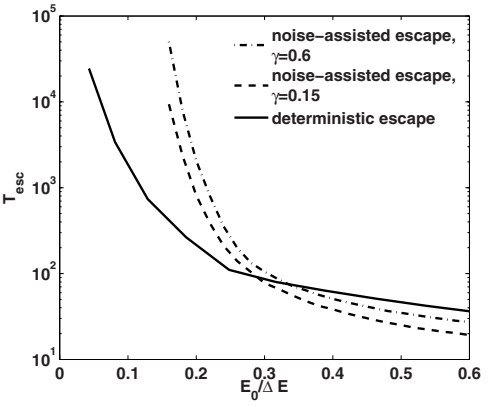

Fig. 5. Mean escape time as a function of the ratio $E_0/\Delta E$ (for details see text) for the microcanonical (*solid line*) and Langevin (*dashed line* and *dashed-dotted line*) dynamics respectively. Parameter values: $N = 100$, $\omega_0^2 = 2$, $a = 1$, $\kappa = 0.3$ and γ as given in the legend.

with growing ratio $E_0/\Delta E$ in the low energy region. This effect weakens gradually for further increasing E_0. Remarkably, for low E_0 the escape proceeds by far faster for the microcanonical system than for the one coupled to a heat bath. For small $k_B T$ the escape time in case of the Langevin system exceeds our simulation time taken as $t = 10^5$ for both depicted values of damping γ.

Concerning the difference between the deterministic and stochastic nature of the formation and stability of the c.l.m. we remark that under microcanonical conditions breather formation proceeds as an inherent and self-organized process. A breather of high enough energy can be created either directly due to a rapidly developing modulational instability or through the subsequent coalescence of smaller-amplitude breathers. In particular, breathers, as coherent structures sustained by the nonlinear chain, are fairly robust, i.e. they are stable with respect to interactions with linear waves. Notably, the deterministic processes take place on a time scale (see above) that is short compared with the time it can take till in the stochastic bath dynamics optimal fluctuations appear that trigger the formation of the c.l.m.. Even if in the stochastic process such a rare event has taken place the formed c.l.m. may readily be destroyed afterwards due to interactions with the heat bath.

5 Conclusions

In conclusion, for the escape of a chain of coupled oscillators from a metastable region over a barrier it is more effective to supply the energy at once and let the system afterwards evolve under microcanonical conditions rather than keeping

the system permanently in contact with a heat bath from which energy can be absorbed. In other form, the underlying deterministic chaotic dynamics, which is generated intrinsically through the interaction of the oscillators propels pronounced energy exchange between the units so that the c.l.m. can be formed on the occurrence of which the escape process is conditioned. At least for small initial energies compared to the barrier values we have found faster transition times. More precisely, while at weak thermal noise the rate of thermal escape is exponentially suppressed, a deterministic nonlinear breather dynamics yields a robust critical nucleus configuration, which in turn causes an enhancement of the noise-free escape rate. Thus, the freezing out of noise may prove advantageous for transport in metastable landscapes, whenever the deterministic escape dynamics can be launched in a single shot via an initial energy supply.

We performed also studies with systems which are more complex than the one-dimensional chain model of harmonically interacting units considered here. It turned out that (i) for chain systems with more than one one degree-of-freedom per unit (e.g. taking into account also motions of the units along the direction transversal to the transition direction) and (ii) for interactions going beyond the linear (harmonic) one (e.g. Morse-type interaction potentials) as well as (iii) other on-site potentials (including biased two-well potentials, potentials being periodic in the direction transversal to the transition direction) the escape time in the microcanonical situation can be significantly shorter than for the corresponding system coupled to a heat bath.

Our study demonstrates the enormous capabilities of nonlinear systems to self-promote their functional processes. Particularly, the ability to manage efficiently (coherent escape) despite being initialized in no ideal condition (far too low energy density compared to the barrier height) distinguishes such systems.

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