COOPERATIVE ESCAPE DYNAMICS OF AN OSCILLATOR CHAIN UNDER MICROCANONICAL CONDITIONS*

D. Hennig, L. Schimansky-Geier

Institut für Physik, Humboldt Universität zu Berlin
Newtonstrasse 15, 12489 Berlin, Germany

(Received October 18, 2006)

We consider the self-organized escape of a chain of coupled oscillators from a metastable state over an energetic barrier. The underlying dynamics is conservative and deterministic. Supply of sufficient total energy or application of external forces brings the chain into the nonlinear regime from which an initially almost uniform lattice state becomes unstable and nonlinear redistribution leads to strong localization of energy. A spontaneously emerging critical localized mode grows to the unstable transition state and the chain, passing through the latter, performs a collective escape process over the barrier. 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.

PACS numbers: 05.40.–a, 05.65.+b, 87.15.–v, 87.15.He

Recently, in the biophysical context there has been a growing interest in barrier crossing of a polymer chain occurring *e.g.* during the transport of long and flexible polymers across membranes, DNA electrophoresis and many other situations [1, 8]. Characteristic velocities of these processes are determined by the corresponding rates of transition out of and waiting times of the chain in the metastable state. Thereby generalizations of Kramers escape theory [9] in over- and underdamped versions have been widely exploited whose first extensions to multi-dimensional systems goes far back to the 60's [10].

While the thermally activated barrier crossing of oscillator chains has been studied extensively [9] less attention has been paid to the zero temperature limit when noise is absent. Here we develop a *microcanonical* sce-

* Presented at the XIX Marian Smoluchowski Symposium on Statistical Physics, Kraków, Poland, May 14–17, 2006.

nario which is based on conserved total energy. The aim of our study is to elucidate the impact of *nonlinear effects* on the escape dynamics in the noiseless situation. We study how a long *discrete* chain of N oscillators induces a transition over an energetic barrier by enhancing one or several localized breather states [11]. It will concentrate the initially almost uniformly distributed energy by internal redistribution without interacting with and gaining additional energy from a thermal bath.

We intend to show that the regime of nonlinear energy localization may promote a faster escape dynamics than imposing the system permanently to stochastic forcing. This is true if the initial energy per oscillator E_0 is small compared to the energetic barrier ΔE. One might compare E_0 with $k_{\mathrm{B}}T$ given to each oscillator in a parabolic potential. In total the chain possesses the energy $E_{\mathrm{total}} = NE_0$. We will show that the nonlinear excitations concentrate a comparably little energy onto a few members which are then able to escape and pull afterwards by kink–antikink motion the remaining chain over the barrier. Therefore, one justly might call this approach a self-organized escape.

The scenario is related with a crossing of a saddle point in configuration space referred to as the transition state [12]. But whereas in a Kramers-like approach the heat bath is sampled for optimal fluctuations to generate the transition state the latter appears in the microcanonical situation spontaneously. This generation is due to a modulational instability [13] of the homogeneous chain in the metastable state and possibly subsequent interaction of breathers.

The *long chain* is modeled as an one-dimensional lattice system of coupled N oscillators with Hamiltonian

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

We assume no bath, and hence no dissipation. The coordinate q_n quantifies the position of the n-th oscillator from a rest value in the direction perpendicular to the axis of the chain. p_n denotes the canonically conjugated momentum. We impose periodic boundary conditions.

The oscillators are coupled harmonically with nearest-neighbor interaction strength κ. Equations of motion read

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

Each oscillator evolves in the anharmonic potential

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

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 $\Delta E = \omega_0^6/(6a^2)$ which oscillators have to overcome individually in order to escape.

In a linear analysis the chain state is expressed as a plane wave solution (phonons) $q_n(t) = A\exp[i(k\,n - \omega t)] + c.c.$ obeying the dispersion relation $\omega^2 = \omega_0^2 + 4\kappa\sin^2(k/2)$ with wave number $k = 2\pi k_0 l/L$. Therein l is the chain spacing, L is the length of the chain (hence it yields $L = lN$) and k_0 are integers in the interval $k_0 \in (-N/2, N/2]$.

But to create breathers the molecular chain has to be brought into a nonlinear regime. The physical situation is that the chain initially is bounded at a certain position which corresponds to a local deep minimum of a potential. In this situation linear forces are effectively acting only and energy equipartition is stable. For the purpose to get energy localization one can imagine two ways:

(i) Sufficient supply increases the energy per oscillator up to a level where nonlinear cooperative excitations become important;

(ii) Application of an external pulling force changes the potential energy such that again intrinsic nonlinear modes are excited.

Importantly these nonlinear modes localize energy and are unable to interact with phonons preventing that the energy gets equalized. In this way the equipartition becomes unstable. Provided there is enough energy the chain will escape from the local minimum due the acting nonlinear forces.

Our physical picture is illustrated in Fig. 1 where we have presented the second scenario to initiate instability of the homogeneous bound state. Let us suppose that for $t < 0$ the chain is located in a stable bound state q_0 (l.h.s.), *i.e.* the energy is almost uniformly distributed in the metastable minimum and nonlinear excitations do not play a role.

For the onset of instability an overcritical force has to be applied. As seen later the inequality (5) has to be obeyed for $t \geq 0$ to get instability. Therefore, the spatially uniform acting force F modifies effectively the parameter a of the potential $U(q)$ of (3). The switch on of the constant force at $t = 0$ modulates the potential in such a way that the relation (5) will hold for all $t \geq 0$. Afterwards, for $t \geq 0$, the parameter a is kept fixed.

The application of the force has initiated a homogeneous elongation of the chain, *i.e.* we have a plane wave with $k = 0$ and $A = q_0$. Small modulational perturbations on this 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

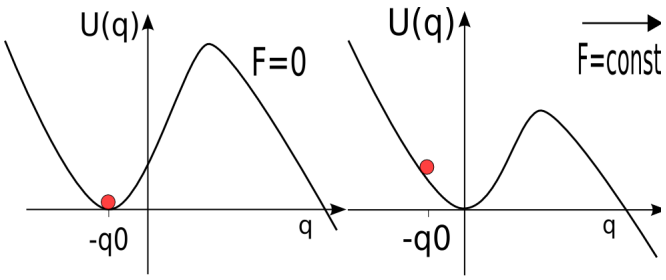

Fig. 1. One-dimensional sketch of the preparation of the chain. Left: For $t < 0$ (l.h.s) the chain (ball) is positioned at the bottom q_0 of a deep local minimum. The equipartition of energy is stable due to the action of linear forces of interaction and of the external potential $U(q)$. Right: We assume the fast turning on of a constant force F acting for $t \geq 0$. This force decreases the barrier and modifies the potential such way (r.h.s) that the chain is now under the influence of nonlinear forces arising from the cubic nonlinearities in $U(q) \propto aq^3$. In result the chain forms breather and equipartition is unstable. This instability let eventually the chain escape with relatively small total energy. Fixing q_0 of the chain during the turning on of the force this position is becoming the relative initial elongation of the chain in response to the action of the external force. We underline that the potential energy gained during this turning on is too small for an escape without nonlinear energy accumulation.

desynchronized ($\Delta q \neq 0$) to have small but nonvanishing initial interaction terms. The energy per oscillator in this situation is small compared to the barrier, *i.e.* $E_0 \ll \Delta E$. Altogether the chain has a total energy of NE_0 plus a tiny but important interaction energy initiating exchange between the coupled oscillators.

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 oscillators.

The exponential growth for $k = 0$ takes place with rate [14]

$$\Gamma = \sin\left(\frac{Q}{2}\right)(8\kappa)^{1/4}\sqrt{\frac{10a^2}{3\omega_0^2}\,q_0^2 - \frac{\kappa}{\sqrt{2}}\,\sin^2\left(\frac{Q}{2}\right)}\,. \tag{4}$$

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

$$a^2 q_0^2 - \frac{3}{10\sqrt{2}} \kappa \omega_0^2 \sin^2\left(\frac{Q}{2}\right) > 0\,, \tag{5}$$

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.

To explore the formation of intrinsically localized modes in our discrete system the set of coupled equations (2) was numerically integrated with a fourth-order Runge–Kutta scheme. The accuracy of the calculation is checked by monitoring the conservation of the total energy. Using as a time step $dt = 0.01$ we find that at the end of our maximal simulation time $t = 10^5$ the accumulated relative error in the conservation of the total energy is less than 10^{-5}. The chain consists of $N = 100$ coupled oscillators.

Starting from an initial flat lattice state of nearly equipartition the attainment of an array of breathers is observed. In more detail, due to modulational instability a pattern evolves in the course of time (of the order of $t \sim 2 \times 10^3$) for which at some lattice sites the amplitudes grow considerably whereas they remain low in the adjacent regions as seen in Fig. 2. The breather states with relatively high energy occur spontaneously at an average distance of the inverse wave numbers $Q_{\max}^{-1}$ corresponding to the maximal growth rate $\Gamma_{\max}$ in (4). If moving these breathers have the tendency to collide inelastically with other ones. 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 strongly concentrated into confined regions of the chain.

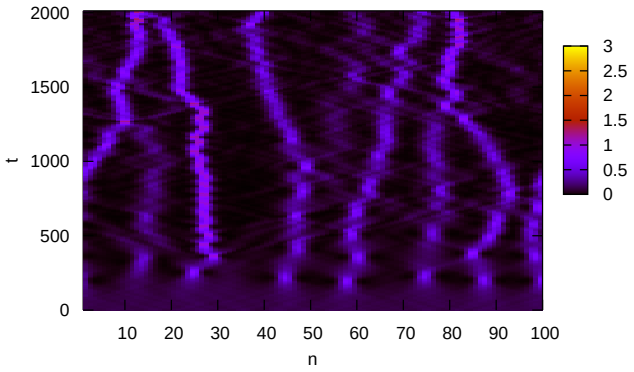

Fig. 2. Density plot of the energy illustrating the formation of a localized structure. The panel on the r.h.s. assigns different colors to the energy content. Initial conditions: $q_0 = 0.3$, $\Delta q = 0.01$, $p_0 = \Delta p = 0$. Parameters $\omega_0^2 = 2$, $a = 1$, and $\kappa = 0.3$. Note the strong energy localization due to breather coalescence around sites $n = 13$ and $n = 81$.

This localization scenario as shown here in case of a metastable potential is characteristic for many nonlinear lattice systems [13, 14]. In the beginning the total energy $E_{\rm total} = 10.936$ which is equivalent to $8.2 \times \Delta E$, is virtually equally shared among all oscillators. The corresponding energy density, *i.e.* the amount of energy contained in a single oscillator, lies significantly below the barrier energy. Precisely, in Fig. 2 the ratio of initial energy and barrier energy is $E_0/\Delta E = 0.08$. In order that a single oscillator escapes it has to accumulate the initial energy content of more than 12 other oscillators.

A breather may even grow such strongly in amplitude that the barrier is surmounted. Whether a oscillator 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 $\tilde{q}_n$, that is the *transition state* separating bounded from unbounded solutions, is determined by $\ddot{\tilde{q}}_n(t) = 0$. Eq. (2) reduces to the stationary system $-\partial U/\partial \tilde{q}_n + \kappa[\tilde{q}_{n+1} + \tilde{q}_{n-1} - 2\tilde{q}_n] = 0$. This equation can be derived from the functional of potential energy $E_{\rm pot}(\tilde{q}_n) = \sum_n \left(U(\tilde{q}_n) + \frac{\kappa}{2}[\tilde{q}_n - \tilde{q}_{n-1}]^2\right)$ yielding in the critical configuration vanishing force as $\partial E_{pot}/\partial \tilde{q}_n = 0$. The corresponding solution of minimal energy is on the lattice chain equivalent to a localized hump solution $\{\hat{q}_n\}$ resembling the form of a hairpin. In Fig. 3 profiles of this *critical localized mode* (c.l.m.) $\{\hat{q}_n\}$ for several coupling strengths are displayed. The stronger the coupling the larger the maximal amplitude of the hump and the wider the spatial extension of the latter. We underline that on a sufficiently extended lattice the c.l.m. represents a narrow state, *viz.* its width is much smaller than the chain length. Apparently a rising coupling κ implies an increase of the *critical energy* $E_{\rm crit} = E_{\rm pot}(\hat{q}_n)$. The latter is needed at least to get

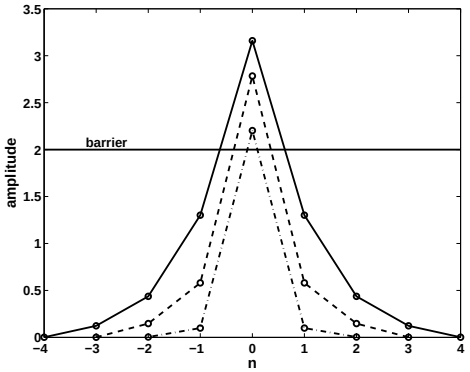

Fig. 3. Amplitude profile of the critical chain configuration for different coupling strengths: $\kappa = 0.1$ (dashed-dotted line), $\kappa = 0.5$ (dashed line), and $\kappa = 1$ (solid line). Note, that for better illustration only a small part of the lattice chain with seizable elongations is shown. Parameters: $\omega_0^2 = 2$, $a = 1$.

the chain elongated into its c.l.m. Since the critical equilibrium solution $\{\hat{q}_n\}$ determines a "force-free" chain configuration it tells us that once the amplitudes have exceeded those of the (unstable) critical equilibrium configuration an overall positive force acts on the oscillators so that they move unidirectionally forward to escape as illustrated in Fig. (4) where the temporal behavior of the force *versus* the lattice position is shown. Hence if the kinetic energy overcomes the critical nucleus subsequent escapes of the neighbors are initiated which progress on the chain to the left and right of the hair pin as a propagating kink and antikink [4,15,16]. In this manner an efficient lowering of potential energy is accomplished. Since kinetic energy of this outward motion is increased return over the barrier into the well is prevented.

The escape time for the chain, defined as the average of the moments at which the N amplitudes of the escaping oscillators pass through a certain value q beyond the barrier (chosen as $q = 10$), as a function of the coupling parameter is displayed in Fig. (5). Progress of escape is crucially influenced by the fastest growing perturbational mode with wave number $Q_{\max} = 2\pi N_{\max} l/L$ determining the number of oscillators, $N_{\max}$, belonging to the arising localized pattern (*cf.* Eq. (4)). The optimal strategy for fast escape is that one of the emerging humps, the number of which is $N_{\text{hump}} = N/N_{\max}$, contains already energy larger than E_{crit}. In this case the localized pattern appropriate for escape has been provided already by the mechanism of modulational instability. Contrary, in case that $N_{\max} E_0 < E_{\text{crit}}$ further energy accumulation due to breather coalescence is required which delays the escape process. To illustrate the impact of the growth rate on the degree of localization of emerging patterns we present in Fig. 6 the energy distribution

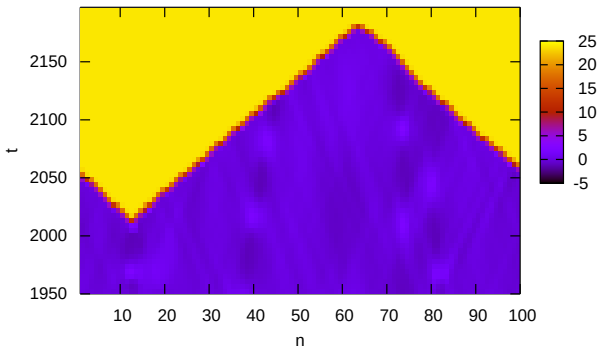

Fig. 4. Escape process: Time-evolution of the force *versus* lattice position. The panel on the r.h.s. attributes different colors to the magnitude of force. Parameters as in Fig. (2).

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

at an early instant of time, namely after the formation of the spatially localized structure due to spontaneous modulational instability has taken place. For comparison patterns for three coupling strengths are shown. In all cases a number of isolated localized humps has formed. The number of humps N_{hump} can be attributed to the wave number at maximal growth rate as follows: $\lambda_{\mathrm{pattern}} N_{\mathrm{hump}} = N$ and $\lambda_{\mathrm{pattern}} = 2\pi/Q_{\max}$. Most importantly, the number of humps (besides their height and width) regulates how the total energy is shared by them. Clearly, for $\kappa = 0.31$ (Fig. 6 (b)) the energy is stronger localized (fewer humps and of higher height) than in the cases of $\kappa = 0.09$ (Fig. 6 (a)) and $\kappa = 1$ (Fig. 6 (c)). Actually, for $\kappa = 0.31$ the units at sites $n = 11$, $n = 30$ and $n = 68$ contain such sufficient amount of energy $E_n > 4/3$ that they overcome the barrier. Thus a localized pattern appropriate for escape has been provided already by the mechanism of modulational instability. In particular no further (major) energy accumulation, which would delay the escape process considerably, is required.

Finally we compare the microcanonical escape process with a corresponding thermally activated process in the Kramers problem [4–7, 9]. The Langevin equations read

$$\frac{d^2 q_n}{dt^2} + \gamma \frac{dq_n}{dt} + \frac{dU}{dq_n} - \kappa \left[q_{n+1} + q_{n-1} - 2q_n \right] + \xi_n(t) = 0 \,. \qquad (7)$$

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

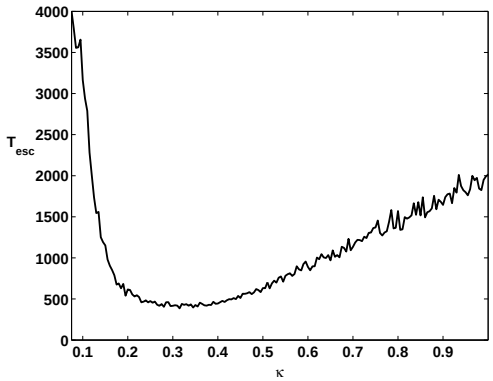

Fig. 5. Mean escape time as a function of the coupling strength κ exhibiting a resonance structure. Remaining parameters as in Fig. (2) except for $q_0 = 0.4$, $\Delta q = 0.1$. Averages were performed over 1000 realizations of initial conditions.

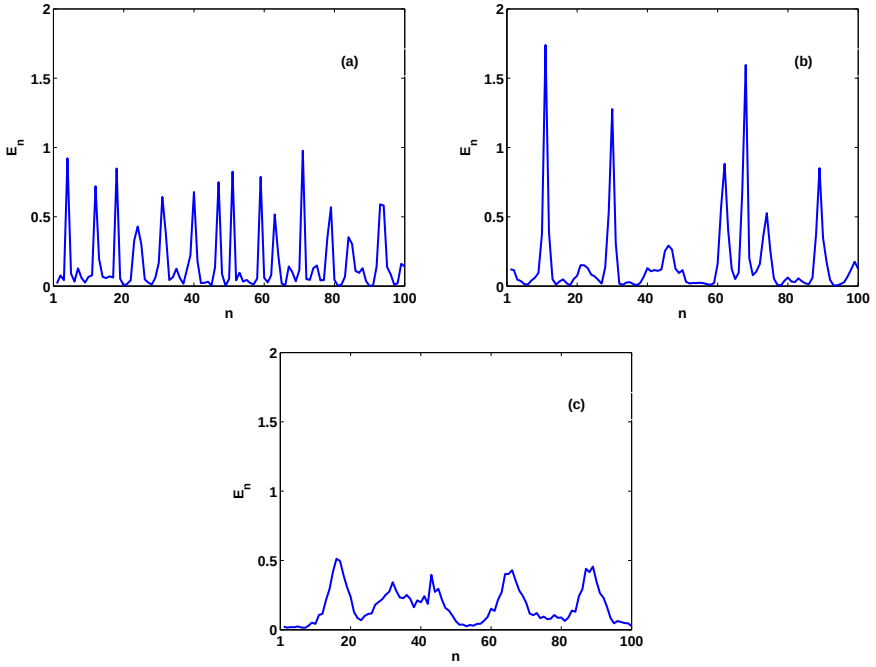

Fig. 6. Spatially localized structure at $t = 500$ for two different coupling strengths. Initial conditions and parameters as in Fig. 5 except for the coupling strength. (a) $\kappa = 0.09$, (b) $\kappa = 0.31$, (b) $\kappa = 1$.

Our results are summarized in Fig. 7 showing the mean escape time of the chain. 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 (2) E_0 is given by the initial energy per oscillator while it corresponds to thermal energy $k_{\mathrm{B}}T$ in case of the Langevin system (7). In both cases there is a rather strong decay of T_{esc} 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 (indicated as the breather region in the plot) the escape proceeds by far faster for the microcanonical system than for the one coupled to a heat bath. For small $k_{\mathrm{B}}T$ the escape time in case of the Langevin system exceeds our simulation time. For $E_0/\Delta E \geq 0.36$ there is crossover so that the mean escape time of the conservative and deterministic system nearly merges with those of the Langevin system. A more detailed study which parameters determine this crossover is in progress.

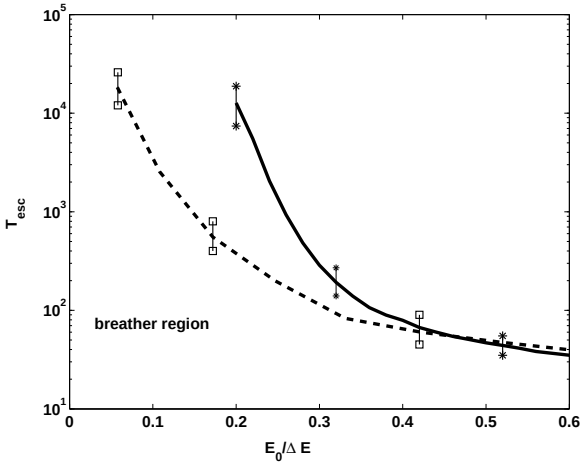

Fig. 7. Mean escape time as a function of the ratio $E_0/\Delta E$ (for details see text) for the microcanonical (dashed line) and Langevin (solid line) dynamics, respectively. Some error bars are shown. Parameters: $\omega_0^2 = 2$, $a = 1$, $\kappa = 0.3$ and $\gamma = 0.1$.

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.

In conclusion, supplying energy to a chain in a metastable state and letting it afterwards evolve under microcanonical conditions is more effective than leaving 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 turns out to provide an efficient mechanism for driving the escape process. At least for small initial energies compared to the barrier values we have found faster transition times. For moderate ratios the differences become negligible.

Applications of this collective escape process are seen for the bound-to-fragmented-state transition of a double-stranded molecular complex, the translocation of trapped one-dimensional polymer through bottlenecks or enzymatic molecular catalysis. Our study demonstrates the enormous capabilities of nonlinear systems to self-promote their functional processes.

We thank for the fruitful discussions with Simon Fugmann and his permanent support. This research was supported by SFB 555 and the VW Foundation Project I/80425.

REFERENCES

[1] W. Sung, P.J. Park, *Phys. Rev. Lett.* **77**, 783 (1996).

[2] P.J. Park, W. Sung, *Phys. Rev.* **E57**, 730 (1998); *J. Chem. Phys.* **108**, 3013 (1998); *J. Chem. Phys.* **111**, 5259 (1999).

[3] D. Kuznetzov, I.E. Dikshtein, L. Schimansky-Geier, *Phys. Rev.* **E** (2001).

[4] K.L. Sebastian, A.K.R. Paul, *Phys. Rev.* **E62**, 927 (2000).

[5] S. Lee, W. Sung, *Phys. Rev.* **E63**, 021115 (2001).

[6] K. Lee, W. Sung, *Phys. Rev.* **E64**, 041801 (2001).

[7] P. Kraikivsky, R. Lipowsky, J. Kiefeld, *Europhys. Lett.* **66**, 763 (2004); *Eur. Phys. J.* **E16**, 319 (2005).

[8] M.T. Dowtown, M.J. Zuckermann, E.M. Craig, M. Plischke, H. Linke, *Phys. Rev.* **E73**, 011909 (2006).

[9] P. Hänggi, P. Talkner, M. Borkovec, *Rev. Mod. Phys.* **62**, 251 (1990).

[10] J.S. Langer, *Ann. Phys. (N.Y.)* **54**, 258 (1969).

[11] S. Flach, C.R. Willis, *Phys. Rep.* **295**, 181 (1998); P. Marquié, J.M. Bilbault, M. Remoissenet, *Phys. Rev.* **E51**, 6127 (1995); H.S. Eisenberg, Y. Silberberg, R. Morandotti, A.R. Boyd, J.S. Aitchison, *Phys. Rev. Lett.* **81**, 3383 (1998).

[12] W. Forst, *Theory of Unimolecular Reactions*, Academic Press, New York 1971.

[13] Yu.S. Kivshar, M. Peyrard, *Phys. Rev.* **A46**, 3198 (1992); T. Dauxois, M. Peyard, *Phys. Rev. Lett.* **70**, 3935 (1993); O. Bang, M. Peyrard, *Phys. Rev.* **E53**, 4143 (1996); M. Peyrard, *Physica D* **119**, 184 (1998); T. Dauxois, S. Ruffo, A. Torcini, *Phys. Rev.* **E56**, R6229 (1997); T. Cretegny, T. Dauxois, S. Ruffo, A. Torcini, *Physica D* **121**, 109 (1998).

[14] I. Daumont, T. Dauxois, M. Peyrard, *Nonlinearity* **10**, 617 (1997).

[15] M. Büttiker, R. Landauer, *Phys. Rev.* **A23**, 1397 (1981).

[16] P. Hänggi, F. Marchesoni, P. Sodano, *Phys. Rev. Lett.* **60**, 2563 (1988).