Parallelization in the modeling of dynamics of a polaron in a constant electric field along molecular chain in Langevin thermostat

Nadezhda Fialko,
Victor D. Lakhno

The Institute of Mathematical Problems of Biology of
Russian Academy of Sciences – the Branch of KIAM RAS

Abstract

- Holstein model of charge transfer in molecular chain (DNA)

  Polaron is the state with lowest energy

- Langevin thermostat

  In a chain without electric field charge distribution (polaron or delocalized state) in thermodynamic equilibrium state depends on the thermal energy of chain

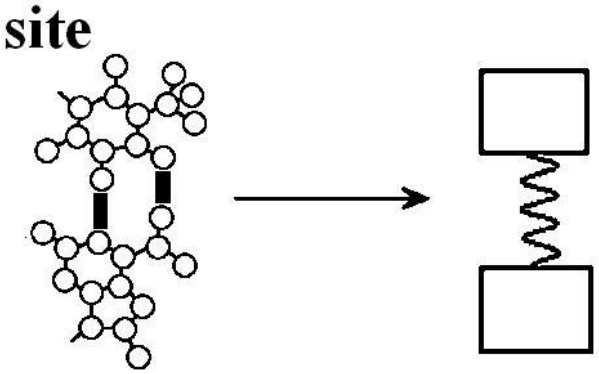
- Simulation of polaron dynamics in a constant electric field

  At fixed T, similar dependencies were found for different-length chains. Polaron mobility was estimated.

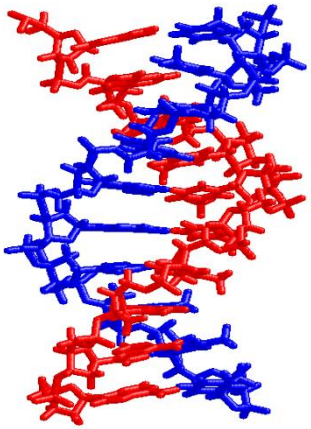
- Variants of parallelization to reduce the calculation time

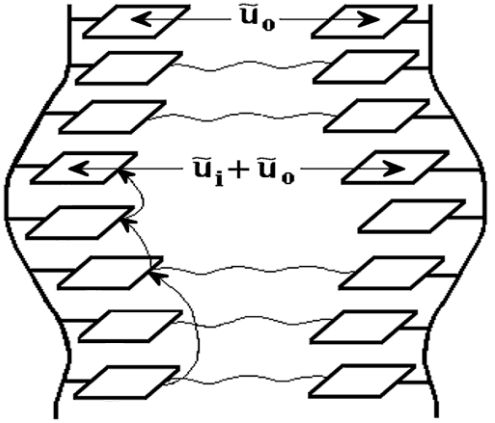
  MPI or openMP

Charge transfer in DNA



Site is complementary nucleotide pair





A quantum particle (electron or hole) moves along a chain of classical sites

The displacements of the sites u_n affect the probabilities of finding a charge $|b_n|^2$

Holstein Hamiltonian

$$\hat{H} = \nu_{nk} |n\rangle\langle k|$$

$$|\Psi\rangle = \sum_n b_n |n\rangle$$

$$H = T + U + \langle \Psi | \hat{H} | \Psi \rangle =$$

$$= \frac{M}{2} \sum_{n=1}^{N} \dot{u}_n^2 + \frac{K}{2} \sum_{n=1}^{N} u_i^2 + \sum_{m=n\pm1} \nu_{mn} b_m b_n^* + \sum_n \nu_{nn} b_n b_n^* + \sum_n \alpha' u_n b_n b_n^*.$$

$b_n(t)$ – amplitude of probability of charge localization on the n-th site

($b_n b_n^* = p_n$ is probability)

$u_n(t)$ – displacements of the center of mass from the equilibrium position

Equations of motion in dimensionless form for a homogeneous chain of *N* sites

$$i\dot{b}_n = \eta(b_{n-1} + b_{n+1}) + \chi u_n b_n + U n b_n$$

$$\ddot{u}_n = -\omega^2 u_n - \chi\,|\,b_n\,|^2 - \gamma\dot{u}_n + \xi Z_n(t)$$

$n = 1,\ldots N$;

η – matrix elements of transition between sites,

ω – frequency of site oscillations,

χ – coupling constant of quantum and classical subsystems,

U – electric field intensity,

γ – friction coefficient,

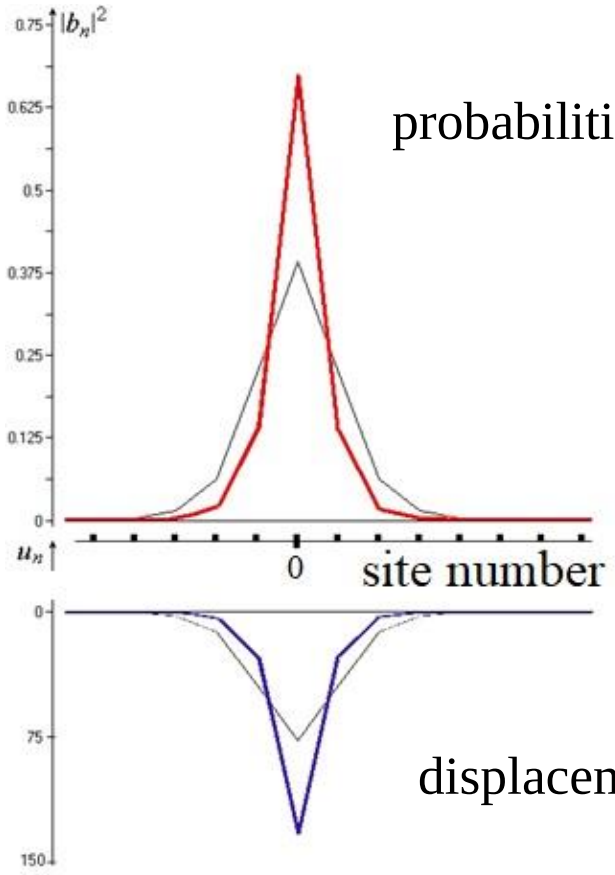
T – temperature,

Z – white noise $\quad \langle Z_n(t)\rangle = 0 \qquad \langle Z_n(t)Z_n(t+t')\rangle = \delta(t') \quad \xi = \sqrt{2E^*}\sqrt{\gamma\,\mathrm{T}}$

System energy

$$E_{\text{tot}}(t) = E_{\eta} + E_{\chi} + E_{\text{Pot}} + E_{\text{Kin}} =$$

$$= \eta \sum (b_n b_{n+1}^* + b_{n+1} b_n^*) + \chi \sum u_n b_n b_n^* + \frac{\omega^2}{2} \sum u_n^2 + \frac{1}{2} \sum v_n^2$$



probabilities $p_n = b_n b_n^*$

displacements u_n

Polaron is the state with the lowest energy

Dynamics of a charge in a constant electric field of intensity *U* along chain at temperature *T*

$$i\dot{b}_n = \eta(b_{n-1} + b_{n+1}) + \chi u_n b_n + Unb_n$$

$$\ddot{u}_n = -\omega^2 u_n - \chi \,|\, b_n \,|^2 - \gamma \dot{u}_n + \xi Z_n(t)$$

For the given *T*, *U*, we compute a set of realizations (dynamics of the system from different initial data and with different pseudo-random time-series) and then calculate trajectories averaged over realizations.

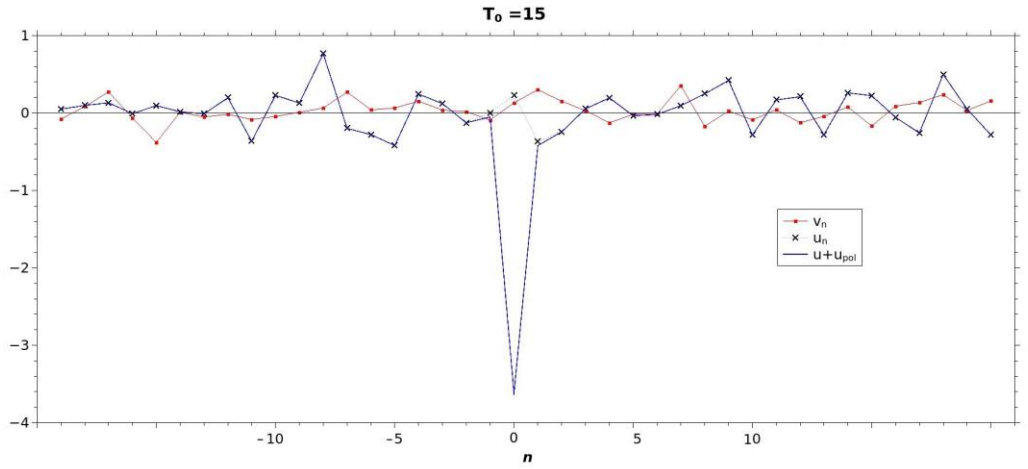
$$b_n(0) = b_{\text{pol}} ,$$

$$u_n(0) = u_{\text{pol}} + u_n ,$$

$$v_n(0) = v_n$$

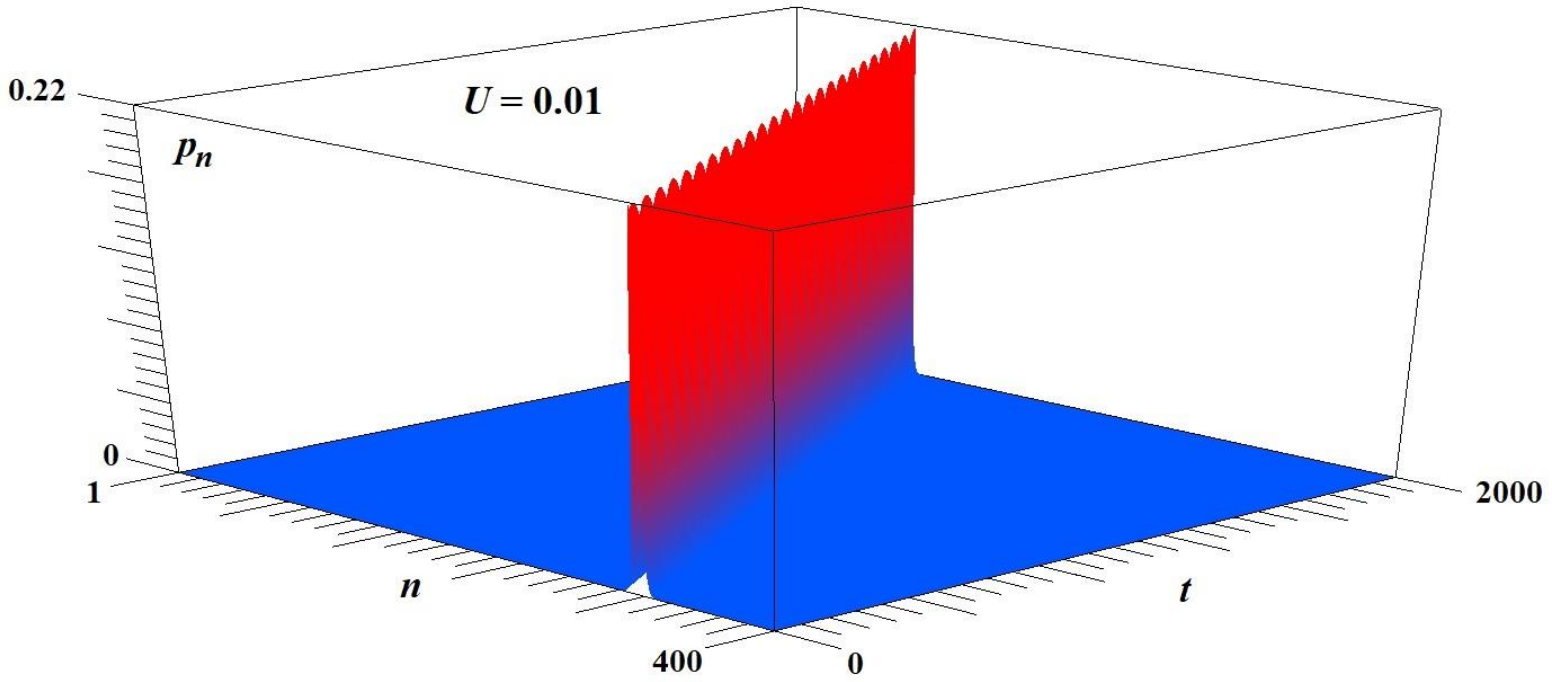
$$\langle u_n \rangle = 0, \quad \langle v_n \rangle = 0$$

$$\langle u_n^2 \rangle = E^* \mathrm{T} / \omega^2, \quad \langle v_n^2 \rangle = E^* \mathrm{T}$$



Initial v_n , u_n and $u_n(0) = u_{\text{pol}} + u_n$ for a realization

Polaron dynamics at T=0

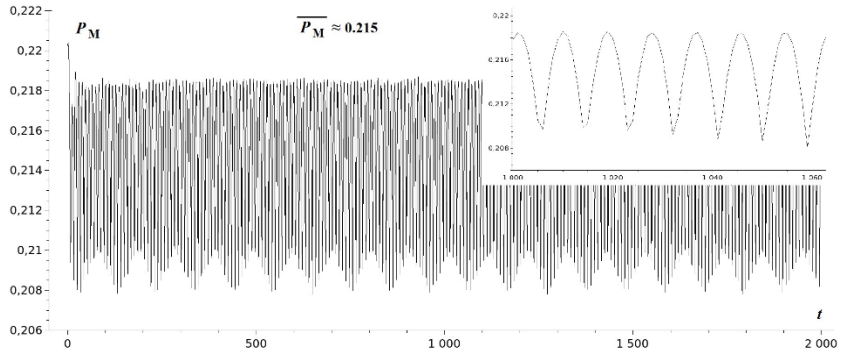


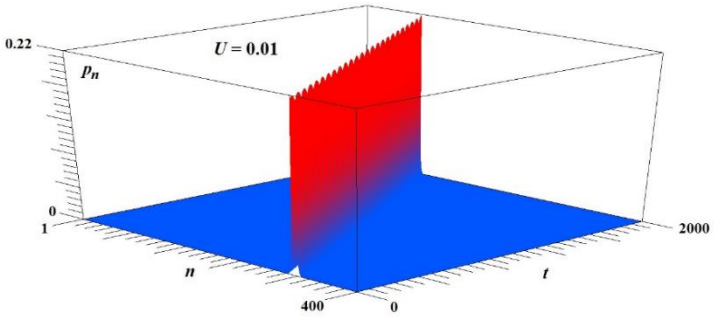
$X(t) = \sum_n (n - n_0) |b_n|^2 = \sum_n (n - n_0) p_n$ displacement of the charge center of mass

charge mobility μ: $\mu U = dX / dt$,

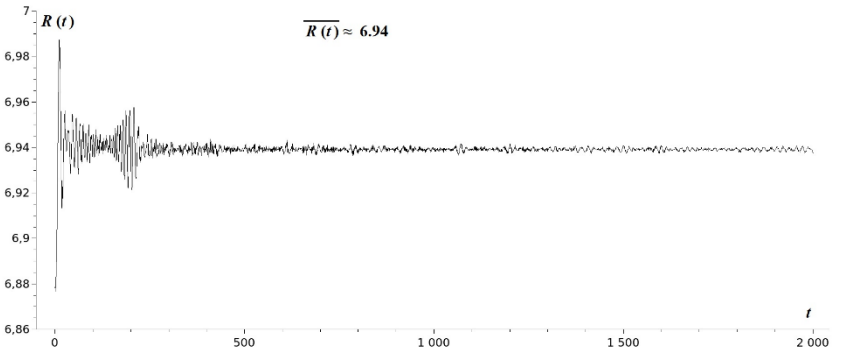
$R(t) = \frac{1}{\sum_n |b_n|^4} = \frac{1}{\sum_n p_n^2}$ parameter of delocalization

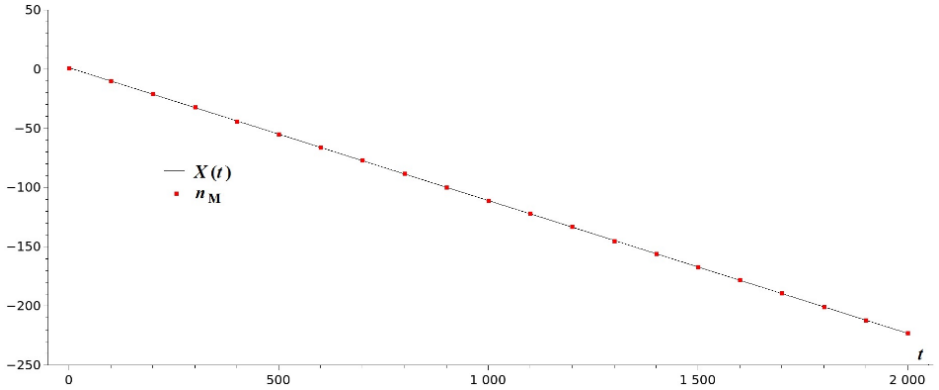
Polaron dynamics (T=0, U=0.01)





$$P_M(t) = \max_n p_n$$





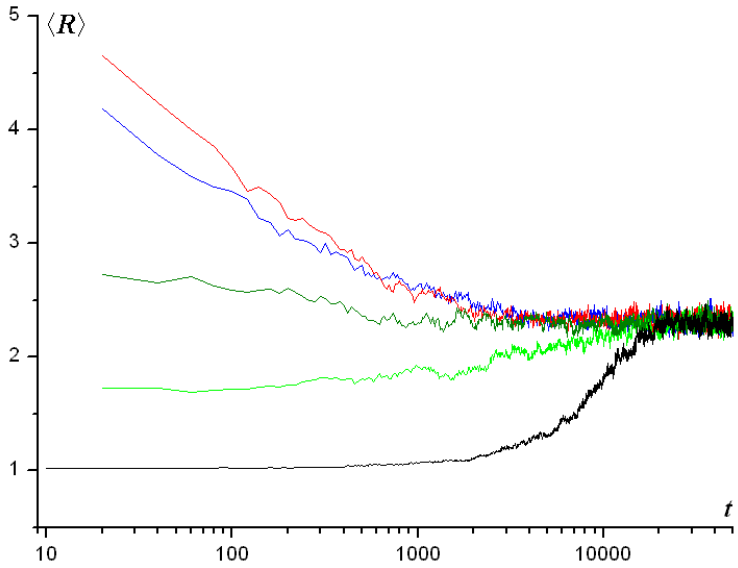
n_M is the number of the site at which P_M is localized

$R(t)$ is almost constant

Graphs $X(t)$ and $n_M(t)$ are the same

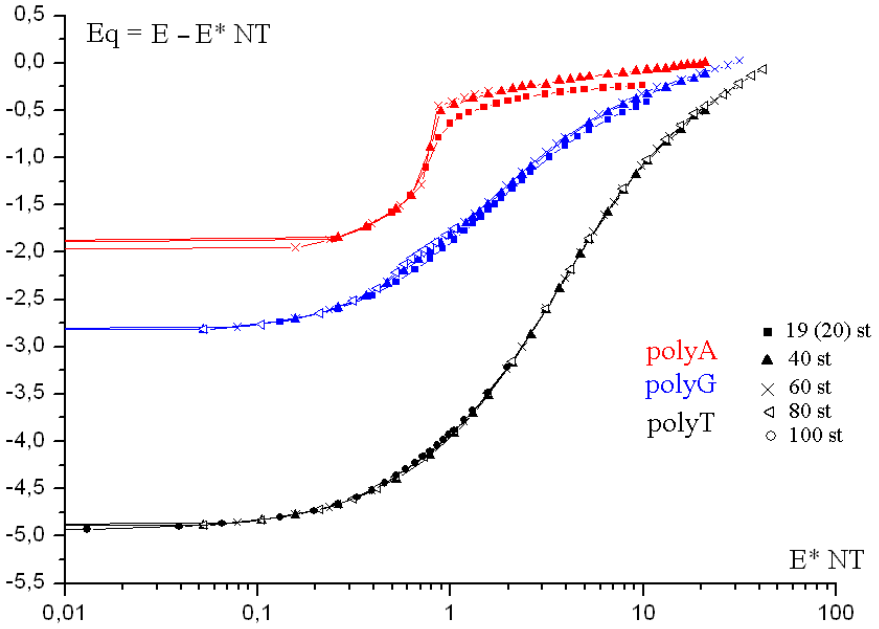
dX / dt does not depend on the chain length N

Polaron dynamics $T \neq 0$, $U=0$



$R(t)$ from different initial data comes to the same state. Black line – initial polaron.

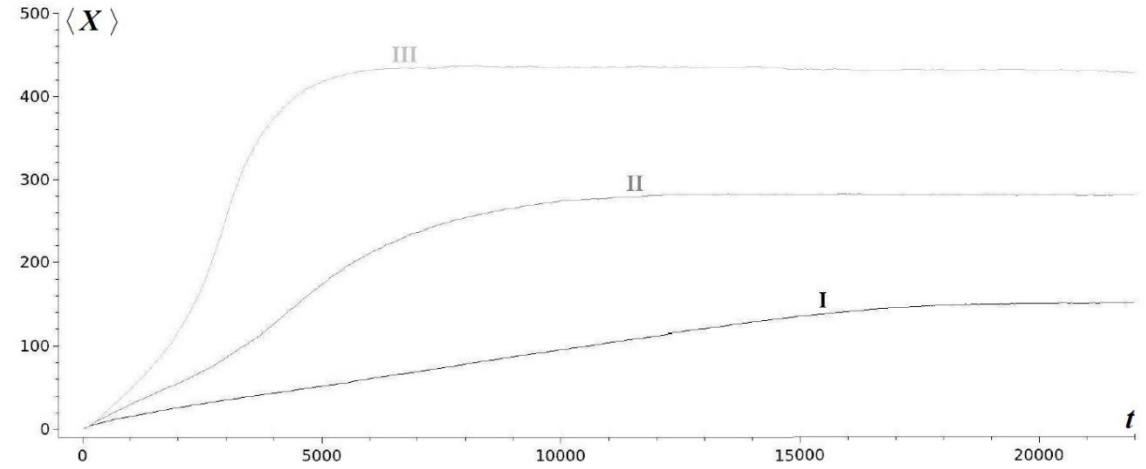
System evoluates from different initial data to similar values $\langle R(t)\rangle, \langle E(t)\rangle$

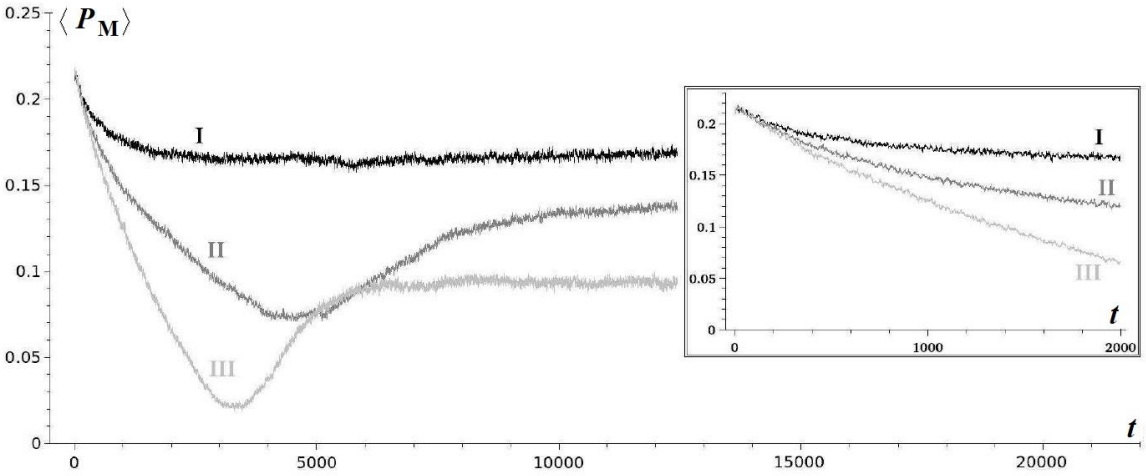


Charge distribution (polaron or delocalized state) in thermodynamic equilibrium state depends on the thermal energy of chain E^*NT

Polaron dynamics $T \neq 0$, $U \neq 0$

Initial polaron existence depend on the thermal energy of chain E^*NT, e.g. on the length of chain N at fixed T.





Chains of
200 sites (I, black),
400 sites (II, gray) and
700 sites (III, light gray).

Each curve is averaged over 100 realizations. The inserts demonstrate the zoomed initial stage.

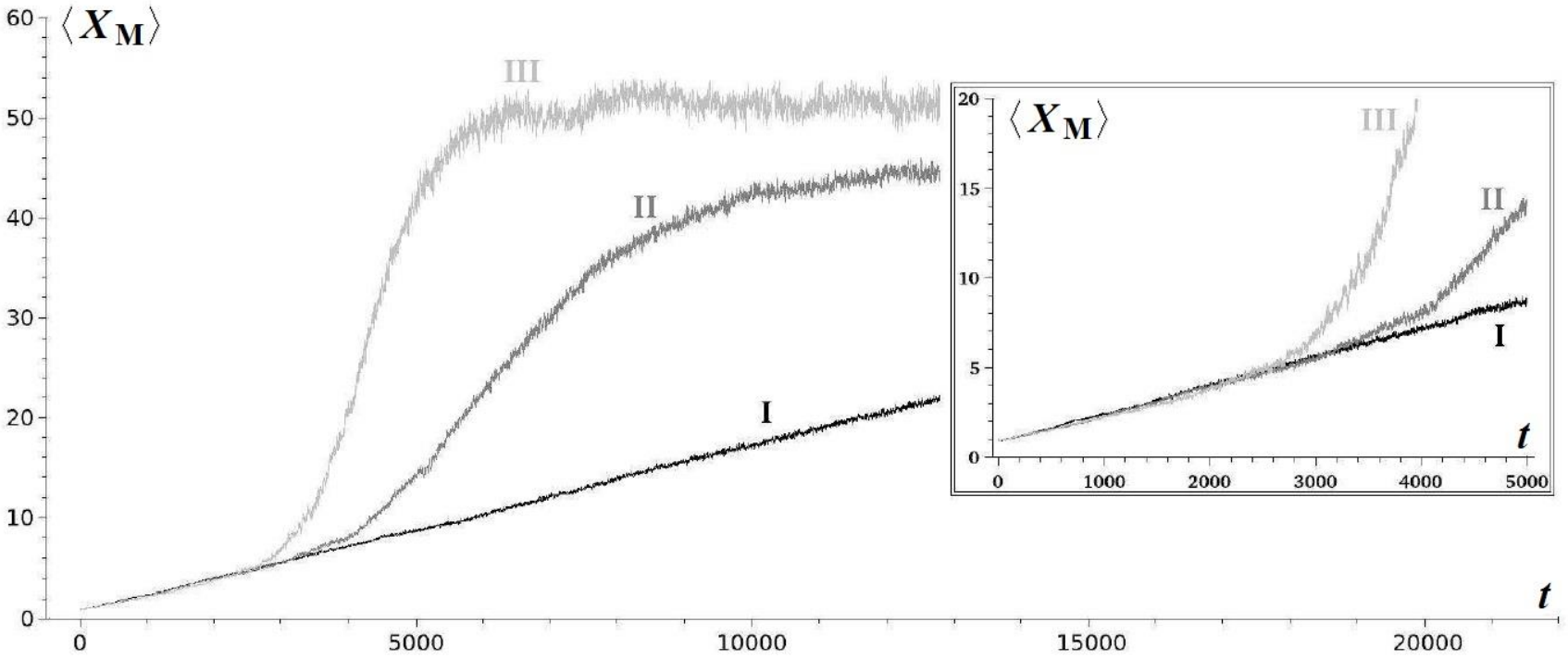
Parameters:
$\eta = 2.4$, $\chi = 1.$, $\omega = 0.5$,
$\gamma = \omega$, $T = 1$, $U = -0.001$

$NT_{crit} \approx 380$

Polaron dynamics $T \neq 0$, $U \neq 0$

$$X_M(t) = P_M(n_M - n_0)$$

analog of $X(t) = \Sigma p_n(n - n_0)$, taking account of only the probability maximum



Chains of 200 sites (I, black), 400 sites (II, gray) and 700 sites (III, light gray). The inserts demonstrate the zoomed initial stage.

The curves $\langle X_M(t) \rangle$ coincide on a march longer time interval, than $\langle X(t) \rangle$.

Estimation of polaron mobility

By analogy with the estimation of the charge mobility μ

$$X(t) = \sum_n (n - n_0) p_n, \quad v = \Delta X / \Delta t, \quad \mu = v / U$$

based on $\langle X_M(t) \rangle$ we estimate the "mean velocity of the polaron" v_M for different U and get the "polaron mobility" μ_M

Polaron velocity v_M and mobility μ_M

T	v_M, $N=200$ $V=0.0005$	v_M, $N=400$ $V=0.001$	v_M, $N=700$ $V=0.005$	$\mu_M = v_M / V$
0.25	0.0012	0.0023	0.011	2.2-2.4
0.5	0.0013	0.0023	0.012	2.3-2.6
1.0	0.0012	0.0024	0.012	2.4

Polaron mobility at $T \to 0$ is very small but nonzero.
For DNA parameter values $\mu_M \approx 0.005$ cm^2/(V·sec).

Computer simulations

$$i\dot{b}_n = \eta(b_{n-1} + b_{n+1}) + \chi u_n b_n + Unb_n$$

$$\ddot{u}_n = -\omega^2 u_n - \chi |b_n|^2 - \gamma \dot{u}_n + \xi Z_n(t)$$

$n = 1,\ldots N$ ($N = 200$, $N = 400$ and $N = 700$)

One realization is trajectory of the system with its own random time-series $\{Z_n(t)\}$ and from its initial data. There exists trivial parallelism “a realization on a core”, using MPI.

Calculations of individual realizations were performed by method similar to Runge–Kutta 2nd order method.

Computation of a realization using openMP

2o2s1g method

$$g_{1,k} = f_k(\{x_n^{(0)} + h^{1/2}\xi Z_n\}),$$

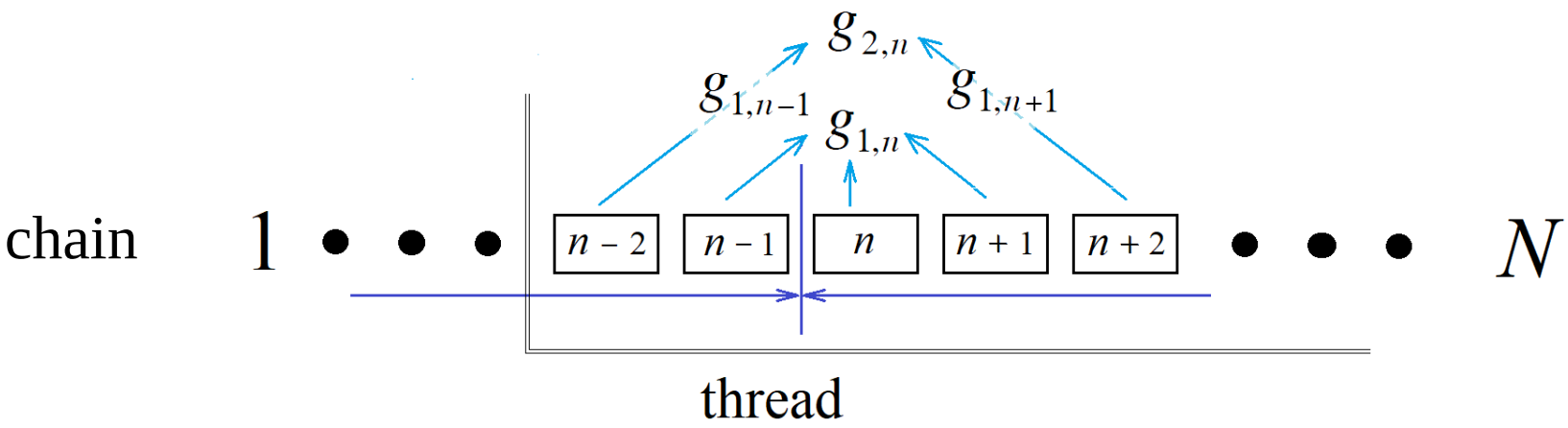
$$g_{2,k} = f_k(\{x_n^{(0)} + hg_{1,n}\}),$$

$$x_k = x_n^{(0)} + \frac{h}{2}(g_{1,k} + g_{2,k}) + h^{1/2}\xi Z_k.$$

$$\dot{b}_n = -i\eta(b_{n-1} + b_{n+1}) + \chi u_n b_n + Unb_n,$$

$$\dot{u}_n = v_n,$$

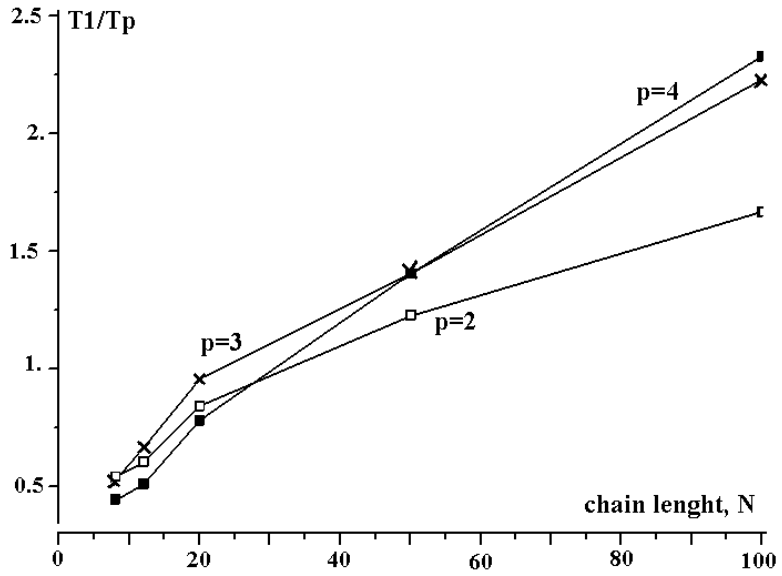
$$\dot{v}_n = -\omega^2 u_n - \chi |b_n|^2 - \gamma v_n + \xi Z_n(t)$$

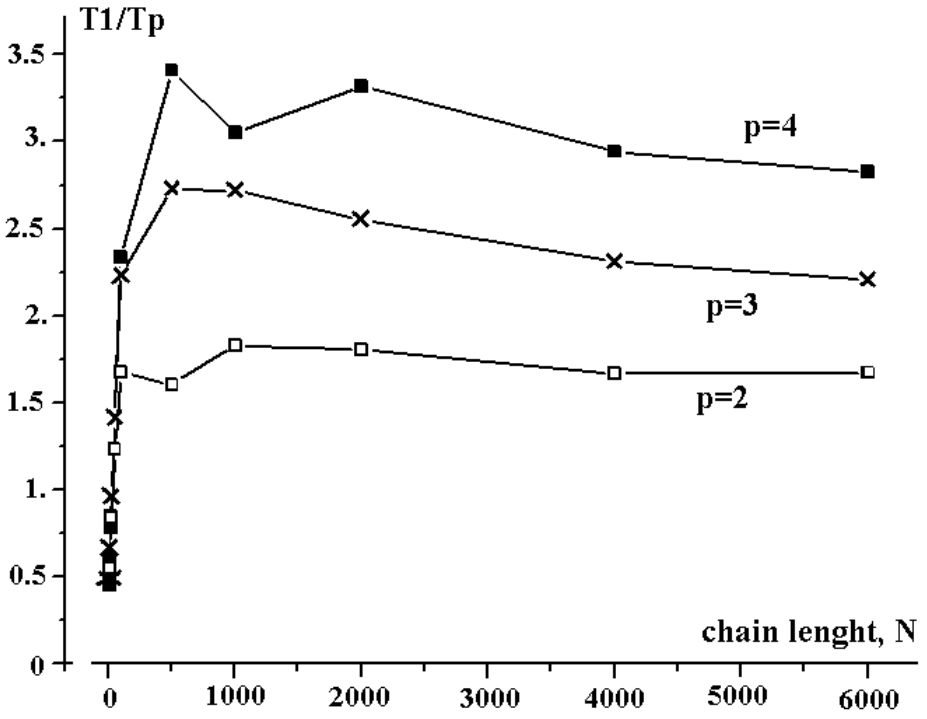


Synchronization is required once per step h

Test results

T1 – machine time of sequentional version,
Tp – duration of the task, parallelized on p threads





Tests were performed on 4-core processors Intel Xeon E5520, Intel Core2 Q9400.

Conclusions

For a large radius Holstain polaron, it is shown numerically that the “mean polaron displacement” behaves similarly for different-length chains. A similar slope of the polaron displacement enables one to find the polaron mean velocity and, by analogy with the charge mobility, assess the “polaron mobility”.

For a given temperature, we compute a set of realizations (dynamics of the system from different initial data and with different pseudo-random time-series) and then calculate trajectories averaged over realizations. This formulation allows for trivial parallelization (using MPI) “one realization – one node” with an efficiency of almost 100%.

To reduce the calculation time of a realization, parallelization was performed on a node containing multi-core processors using openMP. Test studies were made with estimation ratio t1/tp (t1 denotes the execution time of the sequential variant, tp denotes the execution time of the task parallelized using openMP on p threads). Maximum t1/tp ~ 0.8 p allows to significantly reduce the machine time of a realization.

Мы благодарны сотрудникам Суперкомпьютерного Центра коллективного пользования ИПМ им. М.В.Келдыша РАН (http://ckp.kiam.ru)
за предоставленные вычислительные мощности гибридных суперкомпьютеров k-60 и k-100

The calculations were performed on the hybrid supercomputer K60 and K100 installed in the Supercomputer Centre of Collective Usage of KIAM RAS

Thank you for your attension!

Спасибо за внимание!

GRID 2025