# Multiple Time Stepping Methods for Numerical Simulation of Charge Transfer by Mobile Discrete Breathers

Jānis Bajārs[1,a)] and Juan F. R. Archilla[2,b)]

[1]*Faculty of Physics, Mathematics and Optometry, University of Latvia, Jelgavas Street 3, Riga, LV-1004, Latvia.*

[2]*Group of Nonlinear Physics, Universidad de Sevilla, ETSII, Avda Reina Mercedes s/n, 41012-Sevilla, Spain.*

[a)]Corresponding author: janis.bajars@lu.lv

[b)]archilla@us.es

**Abstract.** In this work we propose new structure-preserving multiple time stepping methods for numerical simulation of charge transfer by intrinsic localized modes in nonlinear crystal lattice models. We consider, without loss of generality, one-dimensional crystal lattice models described by classical Hamiltonian dynamics, whereas charge (electron or hole) is modeled as a quantum particle within the tight-binding approximation. Proposed multiple time stepping schemes are based on symplecticity-preserving symmetric splitting methods recently developed by the authors. Originally developed explicit splitting methods do not exactly conserve total charge probability, thus, to improve charge probability conservation and to better resolve high frequency oscillations of the charge in numerical simulations with large time steps we incorporate multiple time stepping approach when solving split charge equations. Improved numerical results with multiple time stepping methods of charge transfer by mobile discrete breathers are demonstrated in a crystal lattice model example.

## INTRODUCTION

Important phenomenon of charge transfer by nonlinear lattice excitations is of particular interest in solid state physics. Recently, transport of charge in silicates by moving nonlinear localized exictations has been experimentally confirmed, a phenomenon known as *hyperconductivity* [1, 2]. Intrinsic localized modes (ILMs), such as *discrete breathers*, *kinks* and *solitons*, have been extensively studied in different crystal lattice models [3, 4]. Commonly, such crystal lattice models at zero temperature are described by classical canonical Hamiltonian dynamics with empirical particle interaction potentials, such as Lennard-Jones and Morse potentials, to name a few, and by thermostated Hamiltonian dynamics, e.g., Langevin dynamics, at a given temperature [5].

Due to different oscillation time scales of charge and lattice particles the transport of charge (electron or hole) by ILMs [4, 6, 7, 8] poses new numerical simulation challenges, which has motivated to consider splitting methods for direct numerical integration of coupled lattice-charge differential equations. Authors in [9] have demonstrated that lattice-charge dynamics can be stated into classical canonical Hamiltonian form and, although the Hamiltonian is not separable in all variables, explicit symplectic and time-reversibility preserving numerical integrators can be constructed with good approximate energy and charge conservation in long-time numerical simulations. This work is stimulated by observations that total charge probability is not exactly conserved by explicit numerical integrators, in contrast to semi-implicit splitting methods (also proposed in [9]), and multiple time stepping schemes, such as the impulse method [5, 10], can be easily incorporated into splitting methods with potentially minimal increase in computational time, thus, ensuring higher accuracy in resolving high frequency oscillations of the charge while performing long-time numerical simulations with large time steps.

## LATTICE-CHARGE HAMILTONIAN DYNAMICS

In this section we describe coupled lattice-charge mathematical model in dimensionless form modeled with the total Hamiltonian $H = H_{lat} + H_c$, i.e., the sum of the lattice and charge Hamiltonians, respectively, where

$$H_{lat} = \quad H_{\mathrm{Q}} + H_{\mathrm{P}} \quad = \sum_{n=1}^{N} \frac{1}{2M_n} p_n^2 + \sum_{n=1}^{N} \left( U(q_n) + \frac{1}{2} \sum_{\substack{n'=1 \\ n' \neq n}}^{N} V(|q_n - q_{n'}|) \right), \tag{1}$$

$$H_c = \quad H_{\mathrm{D}} + H_{\mathrm{A}} + H_{\mathrm{B}} \quad = \frac{1}{2\tau} \sum_{n=1}^{N} \left( a_n^2 + b_n^2 \right) E_n - \frac{1}{2\tau} \sum_{n=1}^{N} \sum_{\substack{n'=1 \\ n' \neq n}}^{N} J(q_n, q_{n'}) b_n b_{n'} - \frac{1}{2\tau} \sum_{n=1}^{N} \sum_{\substack{n'=1 \\ n' \neq n}}^{N} J(q_n, q_{n'}) a_n a_{n'}, \tag{2}$$

and letters Q, P, D, A and B indicate split Hamiltonian terms, which individual Hamiltonian dynamics can be solved exactly with symplectic analytic flow maps $\phi_t^{\mathrm{Q}}$, $\phi_t^{\mathrm{P}}$, $\phi_t^{\mathrm{D}}$, $\phi_t^{\mathrm{A}}$ and $\phi_t^{\mathrm{B}}$, respectively, and is used to construct structure-preserving explicit splitting methods in [9].

In Eq. (1) and (2) $N$ is the number of lattice particles, $q_n \in \mathbb{R}$, $p_n \in \mathbb{R}$ and $M_n > 0$ are the $n^{th}$ particle's position, momentum and rescaled mass, respectively. In the lattice Hamiltonian (1) $U$ is the on-site potential and $V$ is the particle interaction potential energy. For example, such lattice model (1) arises in modeling of a three-dimensional layered crystal, where the on-site potential $U$ models forces from the whole crystal, while the interaction potential $V$ models repulsive and, potentially, as well as attractive forces between particles along a close-packed direction.

Variables $a_n$ and $b_n$ entering the charge Hamiltonian (2) are the real and imaginary parts of $\sqrt{2\tau} c_n$, where $\tau$ is the dimensionless Planck's constant, $c_n$ is the probability amplitude and $P_n = |c_n|^2$ defines the probability of finding an electron or hole at site $n$. The total probability $\sum_{n=1}^{N} P_n = 1$ is conserved as it is the sum $\sum_{n=1}^{N} a_n^2 + b_n^2 = 2\tau$. The introduction of this normalization allows to obtain Hamiltonian dynamics in canonical form [9].

Symmetric charge hopping function $J(q_n, q_{n'}) \geq 0$, i.e., $J(q_n, q_{n'}) = J(q_{n'}, q_n)$, describes charge transfer between states $n$ and $n'$. The transfer function $J$ is often modeled with exponential decay [4, 6] in the following form: $J(q_n, q_{n'}) = J_0 \exp(-\alpha r_{n,n'})$, where $r_{n,n'} = |q_n - q_{n'}|$ and the dimensionless parameter $\alpha > 0$ specifies the rate of exponential decay, while the constant $J_0 \geq 0$ models the probability of charge transfer from one site to another and is model dependent. Smooth function $E_n \in \mathbb{R}$ in (2) describes charge energy at site $n$ and, in general, is dependent on the lattice particle positions, or can be modeled as constant.

Canonical Hamiltonian equations are well known to be symplectic, which implies also phase volume preservation. Apart from the conservation of the total Hamiltonian $H$, the total charge probability above is also conserved. Coupled lattice-charge dynamics is also time-reversible, i.e., Hamiltonian equations are invariant under the transformation $\rho(q, a, -p, -b, -t) = (q, a, p, b, t)$, where $q = (q_1, q_2, \ldots, q_N)^T$, $a = (a_1, a_2, \ldots, a_N)^T$, $p = (p_1, p_2, \ldots, p_N)^T$ and $b = (b_1, b_2, \ldots, b_N)^T$. In addition, equations are invariant under a constant rotation of charge variables $a_n$ and $b_n$, i.e., for any given angle $\theta \in \mathbb{R}$ Hamiltonian equations are invariant under the transformation $\eta(q, \bar{a}, p, \bar{b}, t) = (q, a, p, b, t)$, where $\bar{a} = \cos(\theta) a - \sin(\theta) b$ and $\bar{b} = \sin(\theta) a + \cos(\theta) b$.

The system of canonical Hamiltonian equations derived from $H$ is highly nonlinear and, thus, it is very desirable to obtain an explicit numerical integration scheme, while at the same time preserving as many as possible structural properties of the system stated above.

## MULTIPLE TIME STEPPING METHODS

We propose explicit multiple time stepping methods based on the explicit symplecticity-preserving symmetric method PQDABADQP constructed in [9] with the following numerical flow map

$$\psi_h^{\mathrm{PQDABADQP}} = \phi_{h/2}^{\mathrm{P}} \circ \phi_{h/2}^{\mathrm{Q}} \circ \phi_{h/2}^{\mathrm{D}} \circ \phi_{h/2}^{\mathrm{A}} \circ \phi_h^{\mathrm{B}} \circ \phi_{h/2}^{\mathrm{A}} \circ \phi_{h/2}^{\mathrm{D}} \circ \phi_{h/2}^{\mathrm{Q}} \circ \phi_{h/2}^{\mathrm{P}}, \tag{3}$$

where $h > 0$ is the time step. Notice that the method (3) is constructed from the composition of symplectic analytic flow maps and the symmetry of the method is insured by the *Strang splitting*. Recall that a composition of symplectic flow maps is also symplectic, which is also true for composition of symmetric flow maps, and that symmetric methods are time-reversibility preserving [10]. The method (3) does not exactly conserve Hamiltonian $H$, and neither total charge probability nor rotational invariance, but symplectic methods are known to have very good approximate Hamiltonian conservation properties in long-time numerical simulations [10].

The first multiple time stepping method PQD$\overline{\text{ABA}}$DQP with the numerical flow map reads:

$$\psi_h^{\text{PQD}\overline{\text{ABA}}\text{DQP}} = \phi_{h/2}^{\text{P}} \circ \phi_{h/2}^{\text{Q}} \circ \phi_{h/2}^{\text{D}} \circ \left(\phi_{h/2K}^{\text{A}} \circ \phi_{h/K}^{\text{B}} \circ \phi_{h/2K}^{\text{A}}\right)^K \circ \phi_{h/2}^{\text{D}} \circ \phi_{h/2}^{\text{Q}} \circ \phi_{h/2}^{\text{P}}, \tag{4}$$

where we apply composition $\phi_{h/2K}^{\text{A}} \circ \phi_{h/K}^{\text{B}} \circ \phi_{h/2K}^{\text{A}}$ $K$-times with $K$-times smaller time steps. As the second method we propose also to include piece D into the $K$-times composition, i.e., the numerical flow map of the second multiple time stepping method PQ$\overline{\text{DABAD}}$QP reads:

$$\psi_h^{\text{PQ}\overline{\text{DABAD}}\text{QP}} = \phi_{h/2}^{\text{P}} \circ \phi_{h/2}^{\text{Q}} \circ \left(\phi_{h/2}^{\text{D}} \circ \phi_{h/2K}^{\text{A}} \circ \phi_{h/K}^{\text{B}} \circ \phi_{h/2K}^{\text{A}} \circ \phi_{h/2}^{\text{D}}\right)^K \circ \phi_{h/2}^{\text{Q}} \circ \phi_{h/2}^{\text{P}}. \tag{5}$$

Note that both methods PQD$\overline{\text{ABA}}$DQP and PQ$\overline{\text{DABAD}}$QP are still symlectic, symmetric and second order as the original explicit method PQDABADQP in (3). In addition, during the application of the flow maps $\phi_h^{\text{D}}$, $\phi_h^{\text{A}}$ and $\phi_h^{\text{B}}$ for any $h$ the lattice particle positions $q_n$ do not change, only the momenta values $p_n$ get modified. Thus, both multiple time stepping methods (4)–(5) require only one lattice force and function $E_n$ and $J$ evaluations per time step. Interested readers in explicit representations of all analytic flow maps $\phi_t^{\text{Q}}$, $\phi_t^{\text{P}}$, $\phi_t^{\text{D}}$, $\phi_t^{\text{A}}$ and $\phi_t^{\text{B}}$ are referred to [9].

## NUMERICAL RESULTS

For numerical demonstrations of proposed multiple time stepping methods (4)–(5) we consider crystal lattice example model described in [9]. Example model considers periodic crystal lattice with only fixed close neighbor interactions, i.e., particle $q_n$ interacts only with its two neighboring particles $q_{n-1}$ and $q_{n+1}$, in addition, the charge can hop only from site $n$ to its two neighboring sites $n-1$ and $n+1$, respectively.

In the dimensionless example model, we consider periodic lattice on-site potential and the Lennard-Jones potential for particle interactions, i.e.,

$$U(q_n) = 1 - \cos(2\pi q_n), \qquad V(|q_n - q_{n'}|) = 0.05\left(\left(\frac{1}{|q_n - q_{n'}|}\right)^{12} - 2\left(\frac{1}{|q_n - q_{n'}|}\right)^{6}\right),$$

respectively. We model charge energy function $E_n$ as a nonconstant function

$$E_n(q_n) = U_c(q_n) + E_0, \quad E_0 \in \mathbb{R}, \qquad U_c(q_n) = -\frac{1}{2}\tau(q_n - q_n^0)^2,$$

where $\tau = 10^{-3}$, $q_n^0 = n - 1$ are equilibrium positions of lattice particles and the charge on-site potential $U_c$ is chosen to be a harmonic potential. The charge hopping function $J$ is considered to have exponential decay as described above with parameter values: $\alpha = 15$ and $J_0 = \tau\exp(\alpha)/2$ [9].

We investigate conservation properties of conserved quantities for different values of $E_0/\tau$, which defines high frequency oscillations of the charge. In Fig. 1 we illustrate numerical results for time step value $h = 0.01$, in lattice with $N = 64$ and with $K = 10$ in multiple time stepping methods. In the left plot of Fig. 1 we show maximal relative errors of the Hamiltonian, while in the right plot of Fig. 1 we show maximal relative errors of the total charge probability, over the whole computational time segment $[0, T_{end}]$, where $T_{end} = 100$. Numerical simulations are performed for twenty-five different ratio $E_0/\tau$ values given by the following formula: $E_0^j/\tau = 10^3(1.6^{j-1} - 1)/(1.6^{24} - 1)\tau^{-1}$, $j = 1, \ldots, 25$, and errors are averaged over eleven numerical simulations with different initial conditions using excitation patterns:

$$(p_{n^*-1}, p_{n^*}, p_{n^*+1})^T = \gamma(-1, 2, -1)^T, \quad \gamma \in \mathbb{R}_{\neq 0}, \qquad (a_{n^*}, b_{n^*})^T = \sqrt{\tau}(1, -1)^T, \tag{6}$$

while setting the remaining values of $p_n$, $a_n$ and $b_n$ to zero, and $q_n = q_n^0$, where $\gamma > 0$ leads to a mobile breather solution traveling to the right and $\gamma < 0$ leads to a discrete breather solution traveling to the left. With such initial conditions (6) we are able to generate nonlinear and localized solutions of charge transfer by mobile discrete breathers in our example model. For Fig. 1 results were obtained with $\gamma_j = 0.4 + jh_\gamma$, where $h_\gamma = 0.05$ and $j = 0, \ldots, 10$.

Figure 1 clearly illustrates not only reduction of errors in total charge probability conservation, but in the Hamiltonian errors as well, especially for large values of ratio $E_0/\tau$. Hamiltonian error curves of methods (4)–(5) agree very well with the Hamiltonian error curve of semi-implicit exactly charge conserving splitting methods proposed in [9]. Interestingly, the probability errors of the method (4) has exactly reduced by factor 100 compared to the errors of the method (3), which can be attributed to the second order approximation and $K = 10$, while the method's (5) errors are even smaller, suggesting that it is important to also include piece D in $K$-times composition, compare (4) with (5).

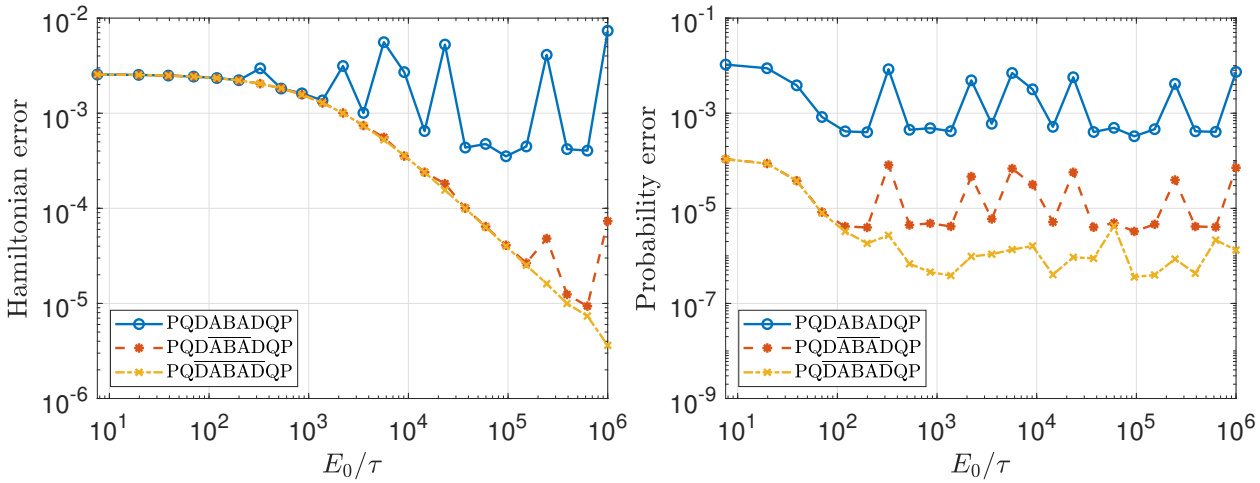


**FIGURE 1.** Hamiltonian (left) and total probability (right) conservation errors of splitting methods PQDABBADQP, PQDABADQP and PQDABADQP averaged over eleven numerical simulations of charge transfer by mobile discrete breathers.

## CONCLUSIONS

In this work we have demonstrated first results of the development of multiple time stepping methods for numerical simulations of charge transfer by mobile discrete breathers in nonlinear crystal lattice models. Results demonstrate that application of multiple time stepping methods improves not only total charge probability conservation, but the Hamiltonian errors as well. Further research is concerned with analysis of choice of value $K$ and obtaining error plots for the Hamiltonian and total charge probability as a function of computational work, and with applications of the multiple time stepping methods to crystal lattice models with realistic potentials.

## ACKNOWLEDGMENTS

J Bajārs acknowledges support from the PostDoc Latvia grant No.1.1.1.2/VIAA/4/20/617 funded by the Latvian Council of Science. JFR Archilla thanks projects MICINN PID2019-109175GB-C22 and Junta de Andalucia US-1380977, and a travel grant from VIIPPITUS 2022.

## REFERENCES

[1] F. M. Russell, J. F. R. Archilla, F. Frutos, and S. Medina-Carrasco, EPL **120**, p. 46001 (2017).
[2] F. M. Russell, M. W. Russell, and J. F. R. Archilla, EPL **127**, p. 16001 (2019).
[3] S. Flach and A. Gorbach, Phys. Rep. **467**, 1–116 (2008).
[4] J. F. R. Archilla, ed., *Quodons in Mica* (Springer, Cham, 2015).
[5] B. Leimkuhler and C. Matthews, *Molecular Dynamics: With Deterministic and Stochastic Numerical Methods* (Springer, Cham, 2015).
[6] O. G. C. Ros, L. Cruzeiro, M. G. Velarde, and W. Ebeling, Eur. Phys. J. B **80**, 545–554 (2011).
[7] L. A. Cisneros-Ake, Physica D **431**, p. 133138 (2022).
[8] L. Cruzeiro-Hansson, J. C. Eilbeck, J. L. Marín, and F. M. Russell, Physica D **142**, 101–112 (2000).
[9] J. Bajārs and J. F. R. Archilla, Mathematics **10** (2022), 3460.
[10] E. Hairer, C. Lubich, and G. Wanner, *Geometric Numerical Integration: Structure-Preserving Algorithms for Ordinary Differential Equations* (Springer Science & Business Media, 2006).