Simulation of energy transport in crystal with NaCl structure assisted by discrete breathers

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Discrete breather (DB) is a spatially localized vibrational mode of large amplitude with vibration frequency outside the phonon band of the crystal. DB frequency can leave phonon spectrum due to the anharmonicity of interatomic bonds owing to the fact that the frequency of a nonlinear oscillator is amplitude dependent. In the case of soft (hard) anharmonicity the nonlinear oscillator frequency decreases (increases) with amplitude. Crystals having a gap in the phonon spectrum can, in principle, support the so-called gap DBs, i.e., DBs with frequencies within the gap. The alkali halide NaI crystal possesses a wide gap in the phonon spectrum and the existence of gap DBs in this crystal has been shown by Kiselev and Sievers with the use of the molecular dynamics method. Later on, several experimental works have been undertaken to support the results of the numerical study and also the possibility of energy exchange between two closely positioned DBs was shown by atomistic simulations. In the present study the energy exchange between DBs in larger clusters is simulated by molecular dynamics. It is shown that the energy initially given to the DB cluster stays in the localized form for a long time (hundreds of DB oscillation periods) even though the energy can travel from one lattice site to another and even polarization of DBs can change. These results contribute to our better understanding of the mechanism of energy localization and transport in crystals.

Keywords: discrete breather, intrinsic localized mode, phonon spectrum, lattice dynamics, nonlinear dynamics, alkali halide crystal, NaCl structure.

Моделирование процесса переноса энергии в кристалле NaCl с помощью дискретных бризеров

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Дискретный бризер (ДБ) представляет собой пространственно локализованную колебательную моду большой амплитуды с частотой колебаний вне фононного спектра кристалла. Частота ДБ может выходить из фононного спектра за счет ангармонизма межатомных связей вследствие того, что частота нелинейного осциллятора зависит от амплитуды. В случае мягкого (жесткого) ангармонизма частота нелинейного осциллятора уменьшается (увеличивается) с ростом амплитуды. Кристаллы, имеющие щель в фононном спектре, могут, в принципе, поддерживать так называемые щелевые ДБ, то есть ДБ с частотами в щели фононного спектра. Щелочно-галоидный кристалл NaI обладает широкой щелью в фононном спектре за счет значительной разницы атомных масс анионов и катионов (атом натрия в 5,5 раз легче атома йода). Существование щелевых ДВ в этом кристалле было показано Киселевым и Сиверсом с использованием метода молекулярной динамики. Позже было проведено несколько экспериментальных работ, чтобы подтвердить результаты численного исследования. Кроме того методом молекулярной динамики была показана возможность обмена энергией между двумя близко расположенными ДБ. В настоящей работе энергетический обмен между ДБ в более крупных кластерах моделируется методом молекулярной динамики. Показано, что энергия, первоначально переданная кластеру ДБ, остается в локализованном виде в течение длительного времени (сотни периодов колебаний ДБ), даже несмотря на то, что энергия может перемещаться от одного узла решетки к другому, и даже поляризация ДБ может измениться. Эти результаты способствуют лучшему пониманию механизма локализации и переноса энергии в кристаллах.

Ключевые слова: дискретный бризер, собственная локализованная мода, фононный спектр, динамика решетки, нелинейная динамика, щелочно-галоидный кристалл, структура NaCl.

1. Introduction

The phenomenon of energy localization in nonlinear lattices in the form of discrete breathers (DBs) was first demonstrated by Dolgov [1] and then in a very famous work by Sievers and Takeno [2]. These works have initiated extensive studies on DBs in model systems as well as in crystals [3–7]. In fact, in real nonlinear lattices DBs are not single-frequency modes and they do not demonstrate exact time-periodicity. Chechin et al. have suggested the concept of quasi-breathers for such objects [8]. DB has a very long lifetime because its frequency lies outside the spectrum of small-amplitude running waves of the lattice. DB frequency, depending on the type of nonlinearity of chemical bonds, can decrease or increase with increase in its amplitude. In the former (latter) case we have soft (hard) type of anharmonicity. Note that soft-type anharmonicity DBs can exist only in crystals featuring gaps (in other words, forbidden zones) in their phonon spectra. Crystals with simple lattices, e.g., pure fcc and bcc metals do not have gaps in the phonon spectra, on the other hand, phonon spectra of crystals with more complex structure may have gaps. In particular, alkali halide crystals with large difference in the atomic weights of anions and cations, e.g., NaI or LiBr have a wide gap in the phonon spectrum. The first investigation of DBs in crystals based on molecular dynamics simulations has been done for NaI [9]. This work has stimulated experimental measurements of crystal spectra to support the existence of DBs [10–12]. In [13] it has been argued that the concentration of large energy DBs at thermal equilibrium in NaI is insufficient for their reliable experimental observation. In [14] DBs in NaI have not been detected.

Molecular dynamics studies on DBs in alkali halide crystals with NaCl structure were continued in the works [15–21]. In particular, in [19] it was shown that two closely placed DBs exchange by their energy quasiperiodically. Note that energy exchange between DBs in graphene has been discussed in the works [22–24]. In the present study the effect of energy exchange between many DBs is investigated by molecular dynamics.

2. Simulation setup

We consider a model of crystal with NaCl structure having lattice parameter a. This structure consists of two fcc lattices shifted relative to each other by the vector $(a/2,0,0)$, at the points of one of them light atoms are located, and the points of the other one are occupied by heavy atoms, as shown in Fig. 1a. Each of the atoms has eight neighbors of the opposite type, located on the tops of the regular octahedron. Each cubic translational cell consists of four light and four heavy atoms. The interaction of atoms is described by pair potentials that take into account Coulomb interactions, Born-Mayor repulsion and dispersion interaction [15]. The atomic mass of the light (heavy) component was assumed to be 10 (100) g/mol. The computational cell with superimposed periodic boundary conditions includes 14×14×14 cubic periodic cells of the crystal. Each cell contains 8 atoms, so that the total number of atoms in the computational cell is equal to 21952.
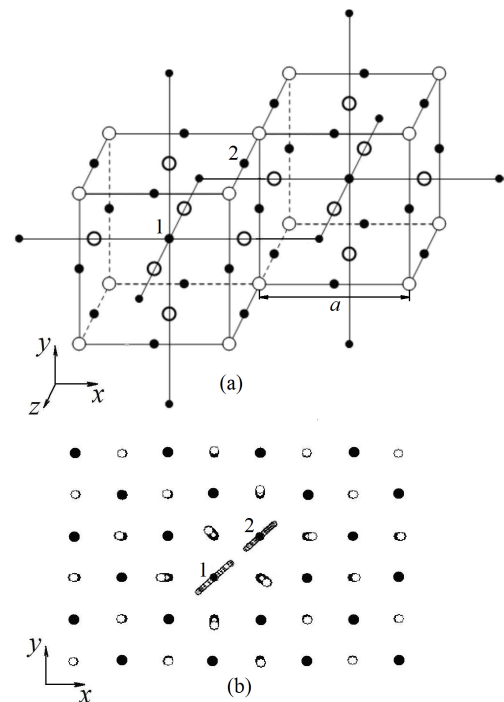
The DB pair was excited by shifting the atoms 1 and 2 from their equilibrium positions along the vectors (−0.3, −0.3, 0) and (0.3, 0.3, 0), respectively. Vector components are given in angstrom. Other atoms initially are in their equilibrium positions. Initials velocity of all atoms is zero.

3. Simulation results

Firstly, for the sake of clarity, we reproduce the results reported in [15,19]. Phonon DOS of the crystal is depicted in Fig. 2a. One can see a wide gap in the phonon spectrum, which appears due to the big difference in the atomic weights of anions and cations. DB in the considered model crystal is highly localized on a single light atom. The atom can vibrate in one of the high-symmetry directions, [001], [110], or [111]. DBs with atomic vibrations along [001] and [011] can be easily excited by the initial displacements of light atoms, but in the case of [111] polarization a more complicated procedure was developed in [20] because the simple method does not work. In Fig. 2b [011] DB frequency is shown as the function of its amplitude. It lies within the phonon gap and decreases with increase in DB amplitude.

The dynamics of DB pair excited on the sites 1 and 2 [denoted in Fig. 1a] is presented in Fig. 3a and 3b, respectively. In this case DBs oscillate out-of-phase along [110] direction, as it can be seen from Fig. 3c, where the phase difference of oscillation of atoms 1 and 2 is shown. Note that time is normalized to the DB oscillation period Θ. Displacement component along z axis is nearly equal to zero for these two atoms.

In Fig. 4 we plot the same as in Fig. 3, but for the case when DBs 1 and 2 oscillate with the phase difference different

Fig. 1. (a) Crystal structure of NsCl type. Light (heavy) atoms are shown by filled (open) symbols. Lattice parameter is equal to a. The fcc anion and cation sublattices are shifted with respect to each other by the vector $(a/2,0,0)$. (b) stroboscopic picture of atomic motion showing the two DBs excited at the sites 1 and 2 shown in (a). The two light atoms vibrate out-of-phase along [110] direction.

from π. In this case quasi-periodic energy exchange between two DBs can be clearly seen. This DB pair was excited by applying initial shifts (−0.31, −0.31, 0) and (0.29, 0.29, 0), in angstrom, to the atoms 1 and 2, respectively.

Our next step is to analyze dynamics of a larger number of interacting DBs. In Fig. 5, 10 light atoms of the chain along [110] direction are labeled as (a), (b), …, (j). Initially two DB pairs were excited on the atoms (a), (b) and (c), (d) by applying the initial shifts (−0.3, −0.3, 0), (0.3, 0.3, 0), (−0.31, −0.31, 0), and (0.29, 0.29, 0), respectively. Vector components are given in angstrom. Note that other atoms had zero initial displacements and all atoms had zero initial velocities.
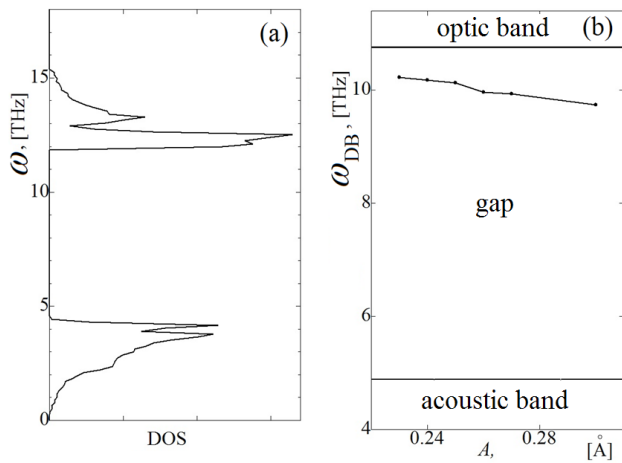
Displacements as the functions of time are shown in Fig. 6a – 6j for the atoms labeled as (a) – (j) in Fig. 5, respectively. It can be seen that initially excited atoms (a), (b), (c), and (d) stop moving after $150t/\Theta$, but their energy

is given to the other light atoms of the chain. Even the atom labeled as (j), which is six sites away from the atom (d), gets excited at about $500t/\Theta$.
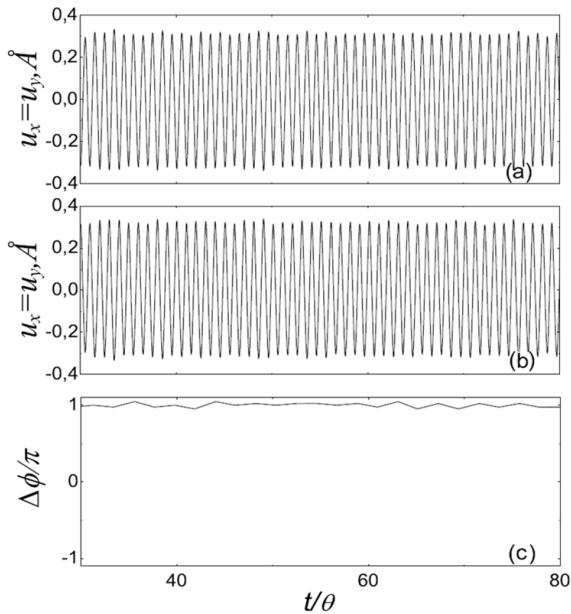
4. Conclusions

From the results presented in Fig. 6 it follows that not only energy exchange between excited DBs in DB clusters is possible but excitation of DBs on the atoms which were not initially excited can be observed. Thus, a new mechanism of energy transport through the excitation of new DBs in expense of the energy of the existing DBs is revealed in the present study.
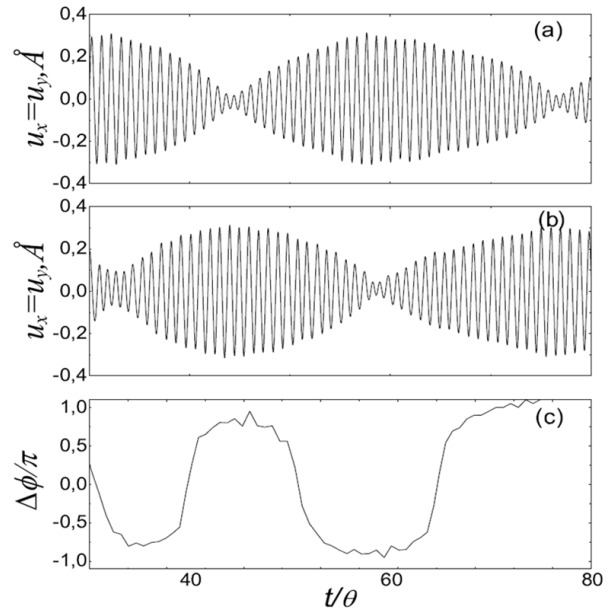
In the future studies it would be interesting to check if DBs can change macroscopic properties of crystals [X,Y] [25,26].
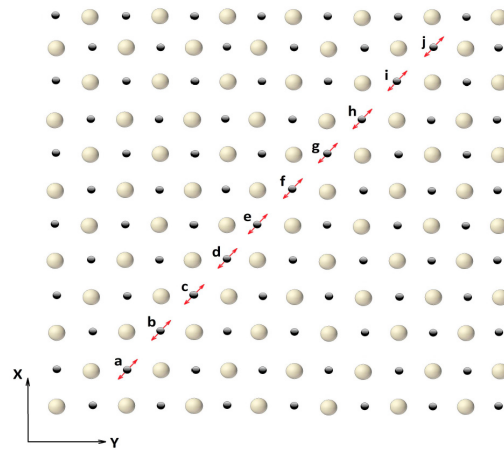

Fig. 2. (a) Phonon DOS of the considered model crystal. A wide gap, which appears due to the big difference in the atomic weights of anions and cations, can be seen. (b) DB frequency as the function of amplitude.
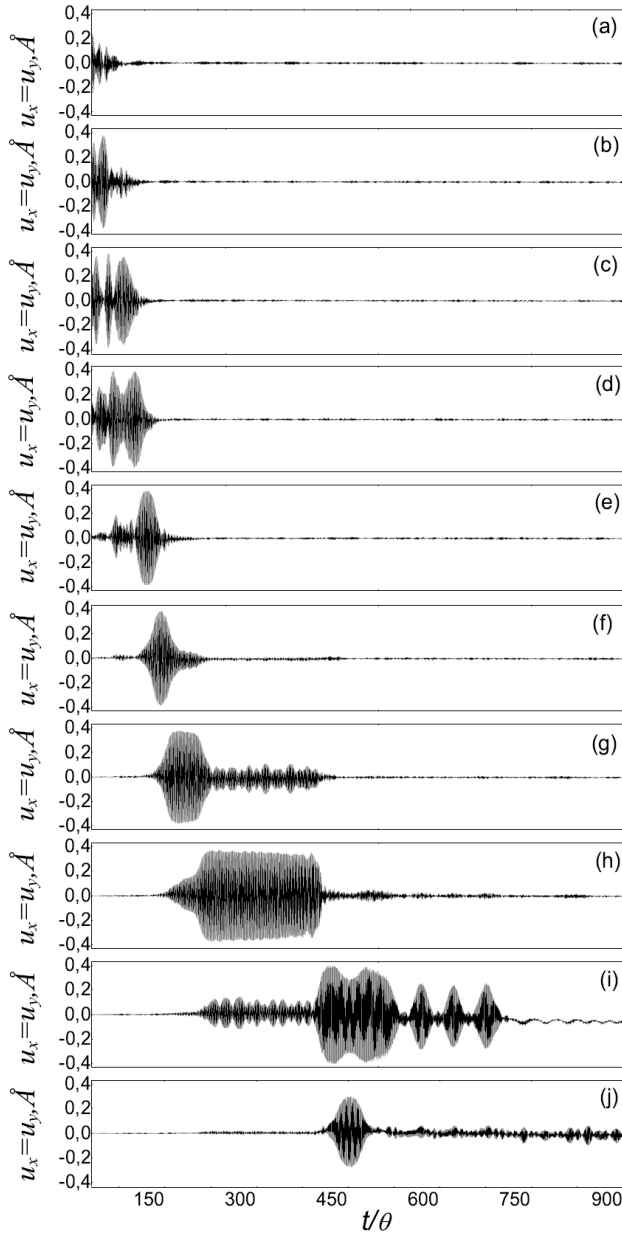

Fig. 4. Same as in Fig. 3 but for the case when atoms 1 and 2 vibrate with the phase difference different from π, and it changes quasiperiodically from $-\pi$ to π and this change is correlated with the energy exchange between DBs.

Fig. 3. (a), (b) Displacements of atoms 1 and 2, respectively, [marked in Fig. 1a] as the functions of time normalized to the DB oscillation period Θ. (c) Phase difference for the oscillation of atoms 1 and 2 showing that they vibrate out-of-phase.

Fig. 5. Ten light atoms of the chain along [110] direction labeled as (a), (b), …, (j). Initially two DB pairs are excited on the atoms (a), (b) and (c), (d).

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Fig. 6. Displacements of atoms labeled in Fig. 5 as (a), (b), …,(j) as the functions of time. Initially only atoms (a), (b), (c), and (d) were excited.