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Article in *Russian Physics Journal* · July 2013

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CONDENSED-STATE PHYSICS

DISCRETE BREATHERS IN CRYSTALS WITH THE NaCl STRUCTURE

J. A. Baimova,¹ S. V. Dmitriev,¹ A. A. Kistanov,¹ and A. I. Potekaev²

UDC 539.2

In the recent decade, the spatially localized large-amplitude vibrational modes in defect-free crystals, referred to as discrete breathers (DBs), are intensively investigated in the materials science. This review reports the main results on gap DBs in crystals with the NaCl structure. The experimental proof of their existence in a NaI crystal is described. A number of molecular dynamics simulation results are presented, including new so far unpublished data. The properties of crystals potentially affected by the DBs are discussed.

Keywords: crystal lattice vibrations, phonon spectrum, discrete breather, family of NaCl crystals.

INTRODUCTION

Discrete breathers are large-amplitude excitations localized in space and periodic in time, which are common in non-linear discrete systems [4–10] and in materials science [2, 11]. The term “breather” originates from the English word *breathe*, which indicates their periodicity in time. Let us briefly describe the physical conditions of the existence of DBs in crystals and their peculiarities distinguishing them from other vibrational modes, using the literature data [1–3].

Crystal lattice excitations can be divided into linear (low-amplitude) and non-linear, where the deviations of atoms from their crystal lattice positions are so large that one has to take into account the non-linear interatomic-force component. The linear excitations include, for instance, low-amplitude plane phonon waves and linear modes localized on the crystal-structure defects. An important feature of the linear modes is their independence of each other, which results from the fact that they represent exact solutions to the atomic motion equations linearized with respect to their equilibrium positions, and for the linear equations the solution superposition principle holds. An increase in the excitation amplitude results in the inclusion of non-linear terms of the extension of interatomic forces over displacements, which results in interaction of different vibrational modes with each other.

The reason, for which DBs exist as localized vibrational modes in defect-free crystals and radiate no energy, consists in the fact that their oscillation frequency lies outside the crystal phonon spectrum. The departure of the DB frequency from the phonon spectrum is due to the dependence of the nonlinear oscillator frequency on its amplitude. In the case of rigid-type nonlinearity, the oscillator frequency increases with the amplitude, and in the case of a soft type of nonlinearity the reverse situation is observed. In the crystals with rigid-type nonlinearity, the DB frequency at sufficiently large amplitudes lies above the phonon spectrum. In the soft-type nonlinearity, the DB frequency would enter the gap of the phonon spectrum, so in this case the presence of a gap in the latter is a necessary condition for a DB to exist.

It was shown in [12] that in nonlinear lattices of a dimensionality higher than 1, the DB amplitude generally exceeded a certain threshold value to let its frequency depart from the spectrum. In this connection, the question has to

¹Institute for Metals Superplasticity Problems, RAS, Ufa, Russia, ²V.D. Kuznetsov Siberian Physical Technical Institute at TSU, Tomsk, Russia, e-mail: julia.a.baimova@gmail.com. Translated from *Izvestiya Vysshikh Uchebnykh Zavedenii, Fizika*, No. 2, pp. 60–69, February, 2013. Original article submitted November 16, 2012.

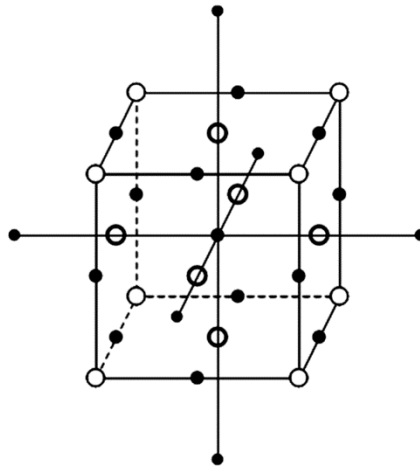


Fig. 1. Crystal of the NaCl structure. Heavy (light) atoms are indicated by open (solid) circles.

be answered as concerns the DB generation mechanisms, since they cannot result from slow energy pumping, starting from the zero level, but only as a result of delivering a certain portion of the energy to a local region of the crystal.

It should be noted that despite a considerable progress in the theoretical description of DBs, the issues of their practical application in the condensed matter physics and materials science have been hardly studied. A certain progress has been achieved in the experimental and theoretical description of DB properties in crystals with the NaCl structure (see Fig. 1). In particular, DBs were experimentally observed in a NaI crystal [4] after their existence had been predicted by the method of molecular dynamics [5]. The purpose of this work is to provide a systematized representation of the well-known data on DBs in the family of crystals under study and to describe new results obtained by the authors, in particular, concerning a possible energy exchange between DBs.

1. EXPERIMENTAL OBSERVATION OF DBs IN A NaI CRYSTAL

Today one of the few experimental proofs of existence of DBs in crystals is their presence in the vibration spectrum of a NaI crystal obtained by the method of inelastic neutron scattering [4]. The experiments were performed at the temperature 555 K. The DB's frequency was close to the bandgap center of the crystal phonon spectrum; DBs were spatially localized and polarized along $\langle 111 \rangle$. It was also shown that the peak intensity in the phonon spectrum, which corresponded to DBs, increased with the temperature, which was expected, taking into account the nonlinear nature of these vibrational modes. In addition, the energy necessary for a DB to appear was estimated to be 0.3 eV. Finally, the authors of [4] arrived at a conclusion that DBs in an alkali halide NaI crystal contribute into the vibrational spectrum of the crystal and can play a significant role in the formation of physical properties at high temperatures.

The experimental investigations of DB's properties in a NaI crystal were continued in [13], where the authors came to a conclusion of an ordered arrangement of discrete breathers in a state of thermal equilibrium. According to the data obtained by neutron scattering, at comparatively low temperatures the polarization of DBs fluctuates between $\langle 011 \rangle$ and $\langle 111 \rangle$. At the temperature 636 K, however, they acquire a stable $\langle 011 \rangle$ polarization, and in so doing the transverse optical mode splits into three modes, which is generally observed during investigations of the superstructures, while the Bragg reflections do not demonstrate any superstructures. The authors argue that observation of a transverse optical mode is associated with ordering of the DB system, which does not disturb the crystal lattice topology.

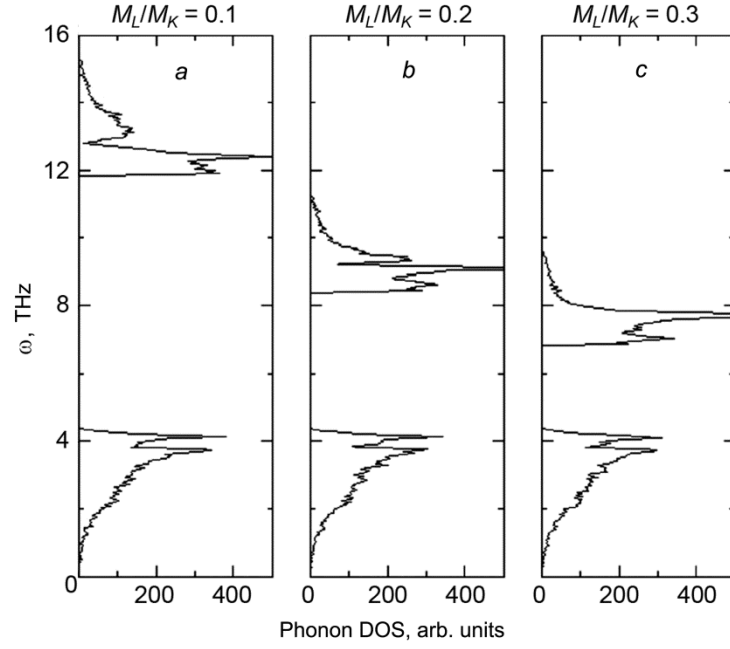


Fig. 2. Densities of phonon states of a crystal of the NaCl structure for the atomic mass ratios of the components $M_L / M_K = 0.1$ (a), 0.2 (b) and 0.3 (c) [8].

2. DATA ON DB MODELLING BY THE METHOD OF MOLECULAR DYNAMICS

2.1. Zero-Temperature Simulations

As mentioned above, the first molecular-dynamics study of DBs in a NaI crystal was made in [5] by Kiselev and Sievers. These authors used real interatomic potentials and a quite complicated algorithm to find a breather solution. They observed a DB of the $\langle 111 \rangle$ polarization and established that its lifetime could be 200 vibration periods and more. It was also shown that this vibration mode was of a soft type of non-linearity, implying that DB's frequency decreases with an increase in the amplitude, splitting from the lower bound of the optical part of the spectrum at the amplitude accounting for 5–6% of the least interatomic spacing.

It was mentioned in the foregoing that DBs with a soft type of non-linearity can only exist, if there is a bandgap in the phonon spectrum. There is no gap in the crystals with a simple lattice; while in the crystals having more than one atom in a primitive translational cell there might be a gap. In the case of multi-component crystals, the gap in the spectrum increases with the difference between the atomic masses of their components.

Figure 2 contains a sample dependence of the density of phonon states in a crystal of the NaCl structure at different ratios of the component atomic masses $M_L / M_K = 0.1, 0.2, 0.3$ [8]. It is evident from the figure that the width of the gap in the phonon spectrum increases as the mass difference is increased.

Even in the case where there is no gap in the phonon spectrum of a crystal, it might appear under the action of elastic deformation resulting in reduced crystal lattice symmetry. On the other hand, it is critical to bear in mind that elastic deformation can result in the formation of a gap in the phonon spectrum of the crystals that have more than one atom in the primitive cell as, for instance, is the case with a graphene [14–16].

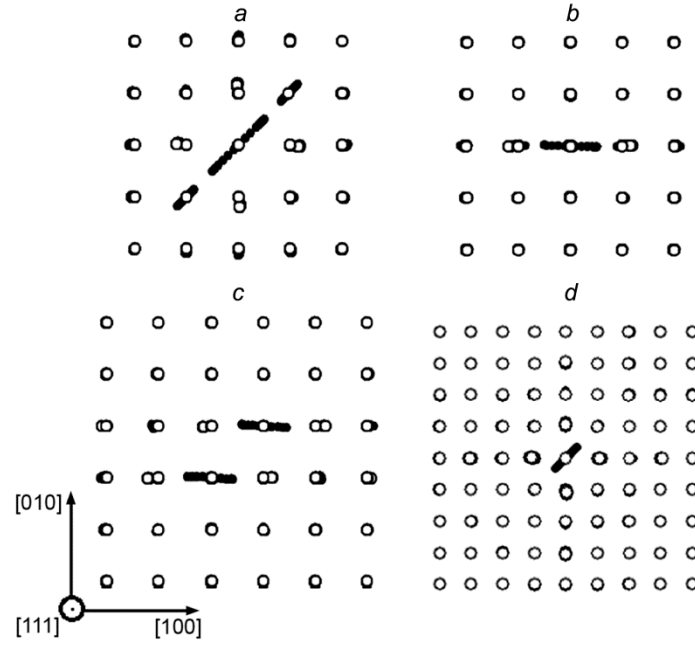


Fig. 3. Stroboscopic images of atomic motion for four types of DBs: an atom with a maximum amplitude vibrates along $\langle 110 \rangle$ (a), $\langle 100 \rangle$ (b), and $\langle 111 \rangle$ directions (d). In (c), two neighboring atoms at a high amplitude vibrate in opposite phases along the $\langle 100 \rangle$ direction. In (a) and (b), the atomic displacements from their equilibrium positions are enlarged by a factor of 7 and in (c) and (d) – by a factor of 5. Heavy (light) atoms are depicted by solid (open) circles.

DBs of different polarizations

In addition to DBs of the $\langle 111 \rangle$ polarization reported in [5, 17], there may exist DBs of the $\langle 100 \rangle$ and $\langle 110 \rangle$ polarizations in crystals of the NaCl structure. Figure 3 presents DBs of different polarizations: an atom with maximum amplitude vibrates along the $\langle 110 \rangle$ (a), $\langle 100 \rangle$ (b) and $\langle 111 \rangle$ directions (d). In (c), two neighboring atoms vibrate in opposite phases along the $\langle 100 \rangle$ direction. In all of the cases observed, light atoms have the highest amplitude and the DB frequency decreases with an increase in the amplitude.

Effect of deformation on DB's phonon spectrum and frequency

Earlier we showed that the type and value of elastic strain considerably affect the width of the gap and the number of these gaps in a crystal of the NaCl structure [18, 19]. DB frequencies in their turn are affected by the compression-tension deformation, but are not so by the shear deformation [19].

Shown in Fig. 4a–c are the densities of phonon states of a crystal for different values of the all-round tension-compression $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon$: $\varepsilon = -0.02$ (a), $\varepsilon = 0$ (b), $\varepsilon = 0.02$ (c), the dependence of the DB $\langle 100 \rangle$ -polarization frequency on ε for a DB with a minimum possible amplitude (d), and the dependence of the DB $\langle 100 \rangle$ -polarization frequency on its amplitude for $\varepsilon = -0.02$ (solid circles) and $\varepsilon = 0.02$ (open circles) (e). The horizontal lines show the upper bound of the phonon-spectrum gap.

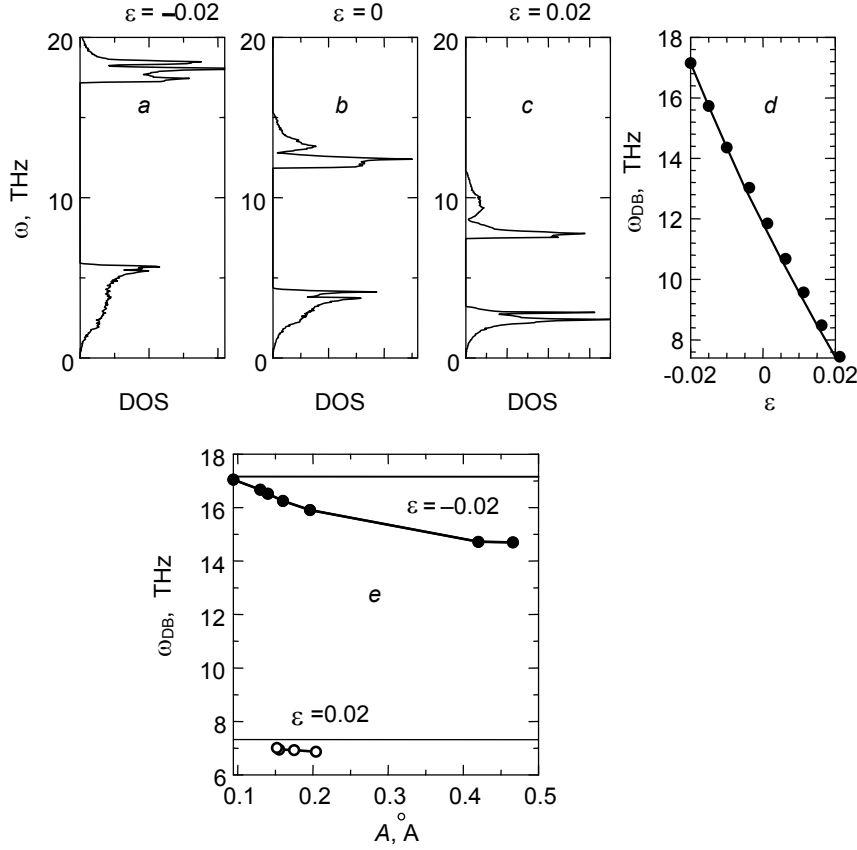


Fig. 4. Dependence of the crystal's phonon spectrum and DB <100>-polarization frequency on all-round tension-compression $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon$ [18, 19]. Densities of phonon states for: $\varepsilon = -0.02$ (a), $\varepsilon = 0$ (b), $\varepsilon = 0.02$ (c), dependence of the DB <100>-polarization frequency on ε for a DB with a minimum possible amplitude (d), and dependence of the DB <100>-polarization frequency on its amplitude for $\varepsilon = -0.02$ (solid circles) and $\varepsilon = 0.02$ (open circles). The horizontal lines show the upper bound of the phonon-spectrum gap (e).

Excitation of DBs in molecular-dynamic calculations

A simplest way to excite a gap DB in the molecular-dynamic calculations is to ascribe a sufficiently high momentum to one of the light atoms at a zero point of time. If the existence of a well-localized DB of a selected polarization is possible, part of the initial-excitation energy would be scattered over the crystal as low-amplitude waves and the other part would remain in a spatially localized form. On the other hand, poorly localized DBs cannot be formed via this method due to the fact that the initial excitation has a pattern far too different from the sought-for vibrational mode. In [5], DBs were excited using a so-called rotating wave approximation.

In the present work we have successfully implemented a simpler but quite effective way of exciting DBs using the swing principle [8, 17–21]. At the zero point of time, one of the atoms is prescribed a *small* momentum and further this atom is affected by an external force, but only in the case where it moves towards its equilibrium. The value of this force is selected in a way to let the amplitude to increase to reach the required magnitude with a few dozens of vibrations. As soon as the prescribed amplitude is achieved, the action of the external force terminates.

A different way of exciting a DB is also possible, i.e., by prescribing a periodic in time motion of a single atom at an amplitude increasing in time to a certain value, following which the system operates on its own [17]. The frequency of this atomic motion would naturally lie outside the phonon spectrum.

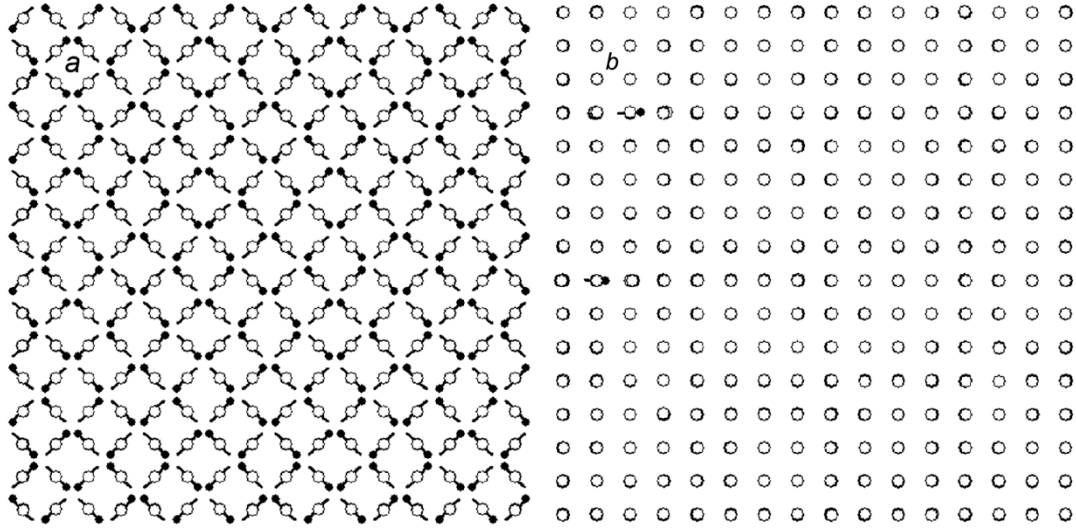


Fig. 5. Short-wave vibrational mode excited in a crystal of the NaCl structure. Atomic displacements are enlarged by a factor of 50 (a). Two $\langle 100 \rangle$ -polarization DBs formed in the crystal as a result of evolution of the modulation-unstable mode presented in (a). Atomic displacements are enlarged by a factor of 10.

The DBs excited in this way would represent stable solutions in crystals at zero temperature. Their stability is implied by the fact that the methods of their construction in numerical calculations contain considerably high perturbations, while the resulting DBs do not practically emit any energy. The presence of thermal lattice vibrations accelerates energy emission by DBs.

In a number of studies, DBs were excited following the mechanism of modulation instability of short-wave phonon modes of considerably high amplitude [8, 22]. Let us describe this mechanism of spontaneous formation of DBs in a crystal of the NaCl structure [8]. A short-wave mode presented in Fig. 5a was excited in this crystal, followed by monitoring of the parameter of light atom motion

$$L = \sum_{n=1}^N E_n^2 \left(\sum_{n=1}^N E_n \right)^{-2}. \quad (1)$$

Here E_n is the total energy of the n -th light atom averaged over the DB vibration period and N is the total number of light atoms in the computational grid. From estimation (1) it is clear that if all of the atoms have an equal energy, then $L = 1/N$, which for large N would be close to 0. On the other hand, if all energy is localized on a single atom, then $L = 1$.

Given in Fig. 6 is the dependence of localization parameter on dimensionless time t/θ for the initial amplitudes of a short-wave vibrational mode 0.02 (a) and 0.03 Å (b). Here θ is the characteristic period of DB vibrations. At the initial point of time, all light atoms possessed equal energy and the localization period was found to be as small as $5 \cdot 10^{-4}$. This low value was maintained for a certain period of time necessary for the short-wave instability to develop. Then the uniform energy distribution was distorted, the localization period was increased reaching a certain level, and further remained nearly constant for a period of time, following which it dropped to a low value. During the time where this parameter is high, a number of DBs persist in the crystal as shown in Fig. 5b. They slowly emit energy in the form of low-amplitude vibrations and finally decay.

Thus the initial state that is far from the thermodynamic equilibrium, where the entire energy is focused in a single vibrational mode, was eventually transformed into an equilibrium state characterized by the energy distribution over all vibrational modes. However, this transformation occurred only after a certain period of a high-degree energy localization in the form of DBs.

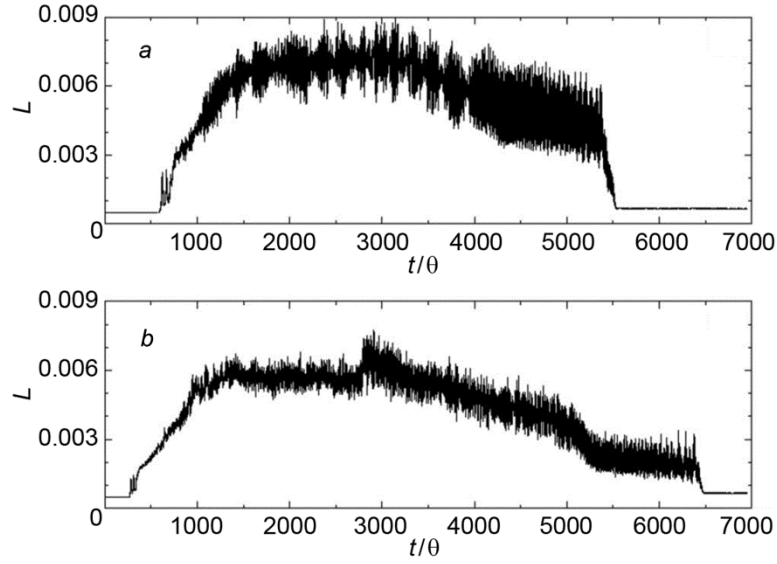


Fig. 6. Dependence of localization parameter (1) on dimensionless time for the initial amplitudes of a short-wave vibrational mode 0.02 Å (*a*) and 0.03 Å (*b*).

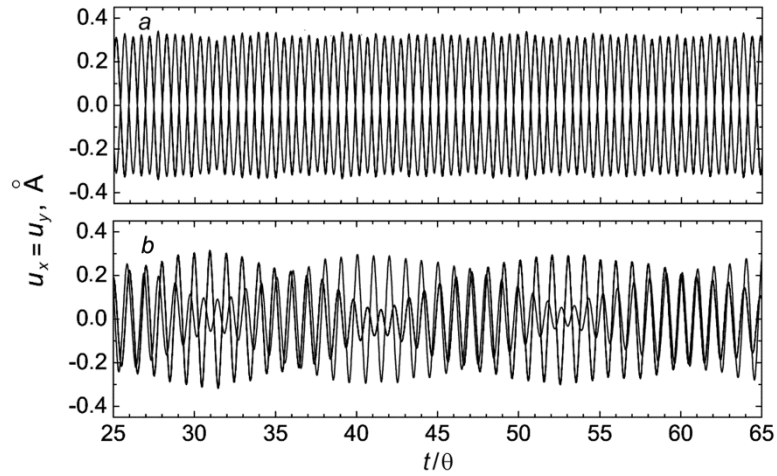


Fig. 7. A pair of $\langle 110 \rangle$ -polarization DBs excited in the opposite phase (*a*) and with a certain phase difference (*b*). Displacements $u_x = u_y$ of two light atoms having a maximum vibration amplitude as a function of dimensionless time t/θ , where θ is the characteristic DB vibration period.

Energy exchange between DBs

In a crystal of the NaCl structure two $\langle 110 \rangle$ -polarization DBs localized on light neighboring atoms were excited along the $\langle 110 \rangle$ direction for the component atomic masses differing by a factor of 10. Shown in Fig. 7 are the displacements $u_x = u_y$ of two light atoms, having the maximum vibration amplitude, as a function of dimensionless time t/θ , where θ is the characteristic DB vibration period. The case of DB vibrations exactly opposite in phase is

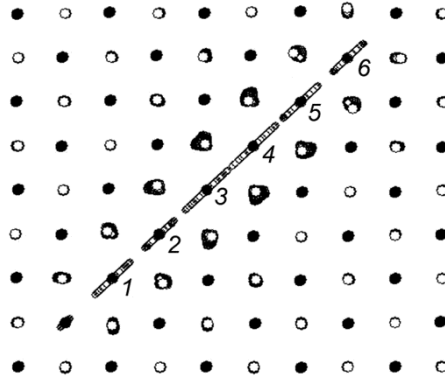


Fig. 8. Stroboscopic image of atomic motion during excitation of four DBs in positions 1, 2, 4 and 5. The DBs in positions 3 and 6 were excited via borrowing energy from the neighboring DBs.

given in (a); while in (b) DBs vibrate with a slight phase difference. It is worth mentioning that in the second case there is an energy exchange between two DBs, so their vibration amplitudes vary periodically.

The energy exchange is possible during excitation of two DB pairs, 1, 2 and 4, 5, which are presented in Fig. 8. It is clear from this stroboscopic pattern of atomic displacements that atoms 3 and 6, which were not excited at the initial point of time, acquired large vibrational amplitude. This implies a possible energy exchange between DBs and a possible propagation of DBs within the crystal via the energy transfer to a neighboring atom.

Figure 9 shows the displacements $u_x = u_y$ of light atoms numbered in Fig. 8 by 1–6, respectively, as a function of dimensionless time. The figure clearly shows the energy exchange between DBs and the energy transfer to a neighboring atom.

Note that the energy exchange between DBs was also investigated for a uniformly deformed graphene [23] and in a one-dimensional non-linear chain [24].

It appears possible to excite complexes of DBs at the frequencies beyond the gap but above the crystal phonon spectrum, which show hard type of linearity rather than soft one. An example of this is given in Fig. 10 showing a u_x displacement for two <100>-polarization DBs adjacent via a heavy atom as a function of time. The DBs have a frequency of 16.5 THz lying higher than the crystal phonon spectrum. Moreover, starting from approximately $t/\theta = 50$, there is a partial energy exchange between DBs.

During excitation of DBs or their complexes in a crystal of the NaCl structure at zero temperature, all kinetic energy appears to be concentrated in a light sub-lattice, since it is the light atoms that vibrate at large amplitude.

2.2. The effect of temperature on DB characteristics

The authors of [25–28] investigated a possible existence of DBs in two-dimensional models of crystals under the conditions of thermodynamic equilibrium at a finite temperature, using simplified interatomic interactions [25–27] and interatomic Morse potentials [28]. It was shown that in the lattices allowing for DBs to exist the lifetime of high-energy atoms increases, which is accounted for by a spontaneous excitation and decay of DBs under the conditions of thermal equilibrium.

For the crystals of the NaCl structure with equal and different masses of anions and cations the density of phonon states was calculated for different temperatures in [29]. It was shown that in a crystal with a considerably large difference between the component masses the phonon state spectrum has a wide forbidden gap, which at considerably high temperatures gives rise to a spontaneous excitation of the gap DBs, whose frequencies lie within the forbidden band. In order to prove this fact, Fig. 11 presents the phonon state densities of the crystal at different temperatures for

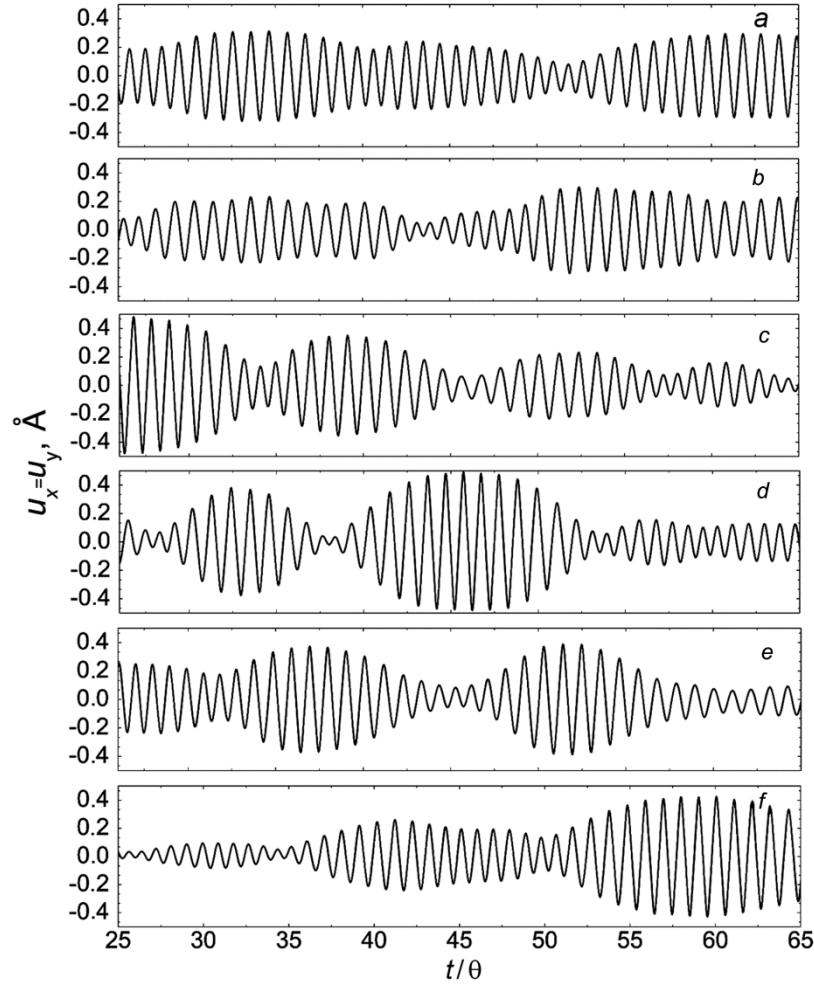


Fig. 9. Displacements of light atoms numbered in Fig. 8 by 1–6, respectively, as a function of dimensionless time.

the cation-to-anion mass ratio equal to 10. It is clear from these data that with an increase in temperature there is a slight decrease in the width of the gap in the phonon spectrum. At higher temperatures (starting from $T = 310$ K) two additional peaks of the density of phonon states are observed, one of which is located in the phonon spectrum gap (around 8 THz, Fig. 11d) and the other lies above the phonon spectrum (about 15 THz, Fig. 11d). The appearance of the peak in the phonon spectrum gap at considerably high temperatures could be attributed to spontaneous excitation of the gap DBs. The formation of a peak above the phonon spectrum is associated with the excitation of DB complexes whose frequencies lie above the phonon spectrum (see Fig. 10). Given this, we can conclude that an increase in the temperature at a considerably large difference between the anion and cation masses in a crystal of the NaCl structure gives rise to increased lifetime and concentration of the gap DBs and their complexes. Similar results were obtained for a two-dimensional, two-component crystal [28].

Note that for an anion mass equal to the cation mass in the crystal of the NaCl structure under study, there is no appreciable influence of temperature on density of phonon states. This is consistent with the fact that the gap DBs in this crystal are not excited due to the absence of a gap in the phonon spectrum.

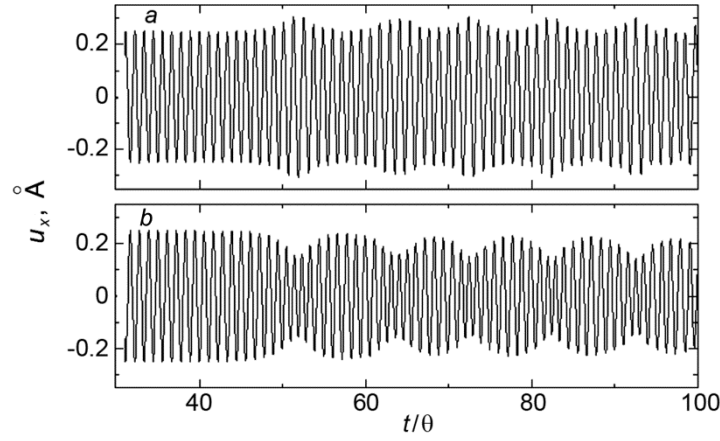


Fig. 10. A pair of $\langle 100 \rangle$ -polarization DBs adjacent via a heavy atom vibrating at the frequency 16.5 THz lying above the crystal's phonon spectrum.

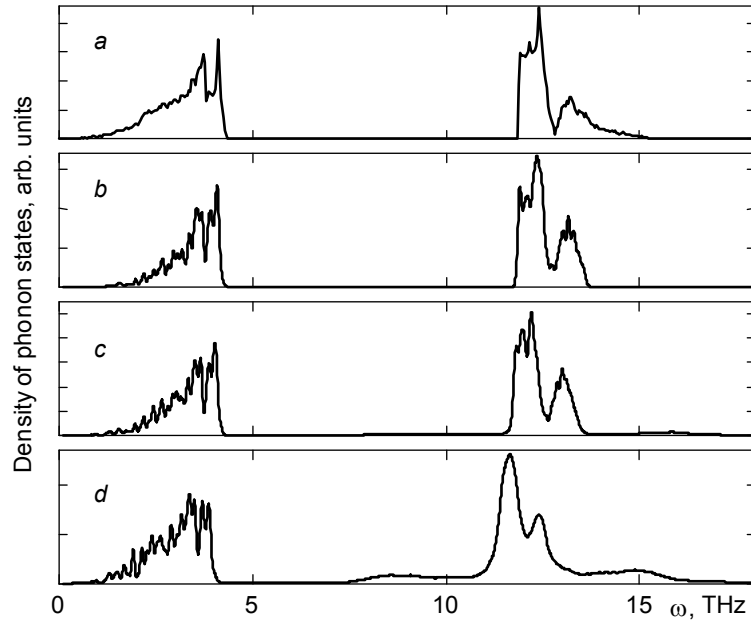


Fig. 11. Density of phonon states of a crystal for the component mass ratio equal to 10 at $T = 0$ (a), 155 (b), 310 (c) and 620 K (d) [29].

SUMMARY

Relying on the DB properties, we can assume which physical properties they would affect and in which phenomena they can play an important role. Firstly, in the crystals having sub-lattices of light and heavy atoms, which accommodate gap DBs, one can expect that an increase in temperature would cause the light sub-lattice to possess a higher temperature than the heavy one. This results from the fact that the DB is localized on light atoms, and an increase in their concentration would result in a relative increase in the temperature of the light sub-lattice. Secondly,

being strongly non-linear vibrational modes, DBs would definitely contribute to the thermal expansion of the crystal [2], since it is associated with the lattice unharmonicity. Being vibrational modes principally different from phonons, DBs have to contribute into thermal capacitance of crystals. DBs are capable of localizing a considerable energy comparable to that of a vacancy [8]. Their possible existence in crystals at finite temperatures and an increase in their energy and lifetime with an increase in the temperature [28] evidences that they can trigger such processes as defect initiation, phase transitions, and other structural rearrangements.

Among the unsolved problems related to the role of DBs in the condensed matter physics and materials science, the following have to be mentioned:

- role of DBs in the thermal equilibrium of crystals and their contribution in thermal capacitance;
- role of DBs in crystals far from thermodynamic equilibrium, for instance, during phase transitions, at high temperature gradients, etc.;
- role of DBs as triggers of structural rearrangements, crystal structure defect formation, and fracture of crystals;
- investigation of electronic structure of DBs;
- possibility of exciting DBs with frequencies lying above the phonon spectrum, i.e., a search for crystals with a hard non-linearity type;
- searching for possible implementations of DBs to solve engineering and technological problems.

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