

CONDENSED-STATE PHYSICS

SPATIALLY LOCALIZED OSCILLATIONS IN LOW-STABILITY STATES OF METAL SYSTEMS

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An analysis of spatially localized and time-periodic vibrational modes (discrete breathers – DBs) is carried out. It is found out that the discreteness and nonlinearity of the medium are the main necessary conditions for the existence of discrete breathers, and these conditions are satisfied in the crystal lattice. The role of DBs in the formation of the physical properties of crystals is discussed. The arguments are presented in favor of the fact that this role increases many times in the states far from thermodynamic equilibrium and in the low-stability pre-transitional states of the crystal lattice.

Keywords: discrete breather, intrinsic localized mode, metal, ordered alloy, nonlinear lattice dynamics, molecular dynamics.

INTRODUCTION

At present, many metallic materials are used in non-equilibrium and low-stability structural-phase states. They function under the conditions of high-intensity external impacts – irradiation, loading, high-density electrical currents, etc. The materials, primarily metallic, are found in pre-transitional states characterized by low-stability of their structure and properties towards external impacts [1–22]. One of possible manifestations of low stability of the crystal lattice consists in the spatially inhomogeneous distribution of the energy and its localization. For these reasons, a discussion of the peculiar behavior of the material structure and properties in the region of pre-transitional low-stability states in the vicinity of the stability threshold attracts close attention. It is in this region of the states, effects, and phenomena in the physical system where one can expect principally new breakthroughs. For the region near the stability threshold, there are no physically grounded concepts, models, approximations, or theoretical approaches so far.

Therefore, in this field the role of structural defects (both static and dynamic) significantly increases [1–22]. A natural structure inhomogeneity results in local inhomogeneity of the spatial energy distribution, primarily the dynamic inhomogeneities in the shape of lattice oscillations [1–22]. Given a low stability of the system towards excitations, the role of these localized crystal lattice oscillations is thought to considerably increase. In essence, such

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dynamic inhomogeneities are the precursors of the structural-phase transformations in the system, with the pathways of the transition channel being quite diverse depending both on the transition pre-history and on the type of a structural inhomogeneity and low-stability state. It was shown earlier that the localized crystal lattice oscillations – discrete breathers (DBs), exist in different types of crystals [e.g., 1, 23, 24]; at present it is important to analyze the peculiarities of DBs in concrete crystals.

The purpose of this study is to analyze the state of the arts in the studies of localized oscillations of crystal lattices of metals and alloys in the form of discrete breathers.

SPECIAL FEATURES OF LOCALIZED OSCILLATIONS – DISCRETE BREATHERS

Let us discuss localized crystal lattice oscillations – discrete breathers, also known as internal vibrational modes. A discrete breather is understood as a spatially localized, time-periodic vibrational mode in a defect-free nonlinear lattice [23]. A discrete breather does not emit its energy, since its frequency lies outside the spectrum of travelling low-amplitude waves (phonons) maintained by the crystal.

Therefore, there are two important questions: the structure of the phonon spectrum of the crystal and the reasons for the DB frequency being beyond the phonon spectrum.

The phonon spectrum of crystals with a simple structure, such as that of pure metals, does not have a gap (frequently referred to as a forbidden band), which implies that such crystals can maintain only DBs with the frequencies higher than that of the phonon spectrum. On the other hand, crystals with complex structure may have a spectrum with a gap; note that they can maintain DBs with the frequencies both higher than those of the spectrum and those lying in the gap (so-called band DBs). The frequency of DBs turns out to be outside the phonon spectrum due to nonlinearity of interatomic interactions, since the oscillation frequency in nonlinear systems often depends on the amplitude [23]. The vibrational mode demonstrates a hard (soft) nonlinearity type in the cases where its frequency increases (decreases) with amplitude. It is evident that DBs with a soft nonlinearity type are possible only in the crystals having a gap in the phonon spectrum [24]. Generally, the frequency of such DBs separates from the upper edge of the gap and sinks into the gap. A gap DB can also demonstrate hard nonlinearity with a frequency separating from the lower edge of the gap, which increases within the gap with the amplitude. In the crystals with a spectrum without a gap only DBs of a hard nonlinearity type can exist [23].

In the discussion of the vibrational mode nonlinearity types (hard or soft) in crystal lattices the benchmark is the study by Kiselev et al. [25] reporting one-dimensional (1D), monoatomic, and diatomic crystals. It is well known that the acoustic and optical parts of the spectrum of a diatomic crystal are separated by a gap (forbidden band). The spectrum of a monoatomic crystal does not contain an optical branch and therefore is gapless.

The investigations were performed for a number of interatomic interactions [25] described by the pair potentials: a polynomial potential $K_2-K_3-K_4$ and by the Toda, Born–Mayer, Lennard–Jones, and Morse potentials. It was shown that the Born–Mayer, Lennard–Jones, and Morse potentials exhibit a soft type of nonlinearity and do not maintain localized modes with the frequencies increasing beyond the phonon spectrum. In a diatomic crystal with a gap in the phonon spectrum it became possible to excite gap DBs with a soft type of nonlinearity. Relying on this finding, a conclusion was made that the lattices with typical interatomic potentials can maintain the vibrational modes with a soft type of nonlinearity only. Therefore, it is very unlikely that DBs would be found in the crystal with a gapless spectrum (such as in pure metals) [25]. No attempts have long been made to excite DBs in pure metals due to the absence of a gap in their spectrum.

It should be noted that the interatomic potentials commonly have an inflection point, wherein the second derivative with respect to distance changes the sign from negative to positive, so that at short distances the potential is hard and at large distances it is soft. This can be represented as follows. The atoms at large distances mainly interact via overlapping of electron clouds, while at short distances the interactions of their nuclei become more important. It can be assumed that if a soft potential tail makes a large contribution into the dynamics of a specific mode, then the mode frequency would decrease with the amplitude, but if there is a prevailing contribution from the hard nucleus, the opposite would be the case.

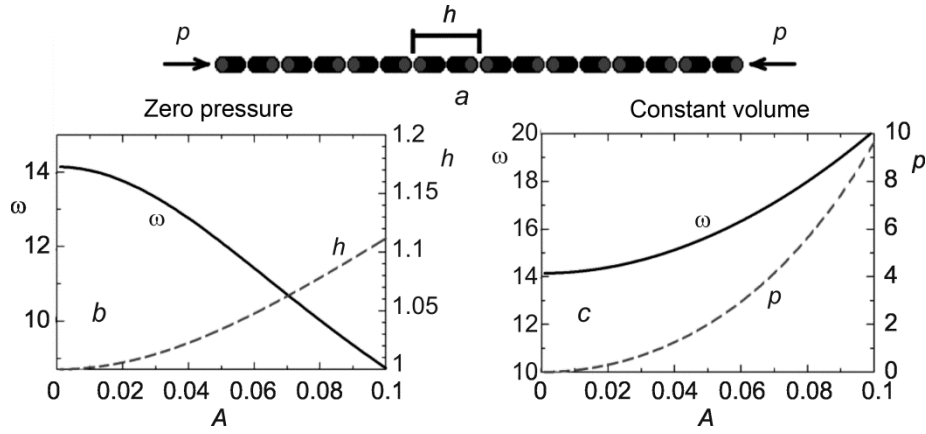


Fig. 1. Short-wave vibrational mode in a one-dimensional monoatomic crystal: all atoms oscillate with the same amplitude A , each of them is in antiphase with the nearest neighbors (a). Frequency (left ordinate) and distance between the nearest atom centers (right ordinate) as a function of A in the case of zero external pressure in the case of a thermal expansion of the chain (b). Frequency (left ordinate) and pressure (right ordinate) as a function of A in the case of constant volume ($h = 1$) in the absence of thermal expansion (c).

Consider a one-dimensional chain of identical unit-mass atoms. Let the interaction of a particle with the nearest neighbors be described by the potential $U = U(r)$. Denote $x_n(t)$ to be the coordinate of particle n , which is an unknown function of time t . Now the chain dynamics is described by the following equations of motion:

$$\frac{d^2 x_n}{dt^2} = U'(x_{n+1} - x_n) - U'(x_n - x_{n-1}). \quad (1)$$

Using this simple model (Fig. 1a), let us discuss the dynamics of a short-wave mode describing the interatomic interactions by the Morse potential. Assume that all particles oscillate with the same amplitude A , every particle being in antiphase with the nearest neighbors. Let us take two cases. In the first case, the interatomic distance h is a function of the mode amplitude A , so the chain is under a zero external pressure. This implies that the chain can undergo free ‘thermal expansion’. In the second case, the distance between the centers of atomic oscillations is fixed (thermal expansion is prohibited), i.e., $h = 1$. As the oscillation amplitude A increases, the chain pressure p would also increase. The mode frequency ω and the interatomic distance h in Fig. 1b are shown as a function of amplitude for the first case. It is clear that ω decreases with increasing A , so the mode in question demonstrates nonlinearity of the soft type. However, in the second case (Fig. 1c) frequency ω and pressure p increase with increasing A . This is due to the fact that, given a constant volume, the hard nucleus of the Morse potential introduces a larger contribution into the mode dynamics, while in the case of zero pressure (where the distance between the atomic vibration centers increases) there is an increasing contribution from the soft potential tail.

In this situation it is clear why a DB cannot exist in a monoatomic chain with an interaction in the form of the Morse potential. Should a spatially localized large-amplitude vibratory mode appear, this would have resulted in local thermal expansion with an increase in the distance between the atomic vibration centers. Moreover, this would have resulted in a larger contribution from the soft potential tail into the mode dynamics, but its frequency could not have increased with the increasing amplitude to be higher than the gapless phonon spectrum.

It has turned out that the results [25] are valid for one-dimensional crystals only. In a two-dimensional lattice with the Morse interatomic potential (Morse lattice) there are various possibilities of suppressing thermal expansion in the DB nucleus, and its frequency can increase with amplitude beyond the phonon spectrum frequency, which makes DB a long-living vibrational mode.

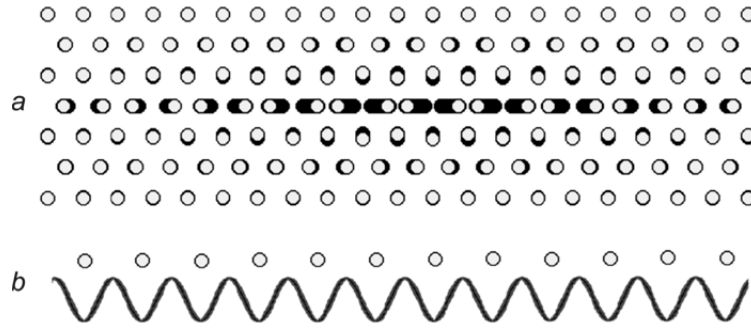


Fig. 2. Rod-like DB in a two-dimensional monoatomic Morse lattice [26] (a). Schematic representation of the atomic chain in the local periodic potential (b).

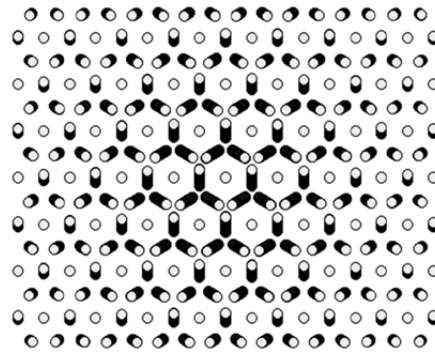


Fig. 3. DB of a radial symmetry in a monoatomic two-dimensional Morse lattice [27].

One possible way of exciting a DB in a two-dimensional Morse lattice is given in Fig. 2. [26]. A rod-like DB is excited in a close-packed atomic row (Fig. 2a). In this DB, about ten atoms of one close-packed row perform large-amplitude vibrations, with their vibration amplitude decreasing with distance from the DB center. It is evident that the atoms in the DB nucleus are deprived of the free thermal expansion, since the atomic row in which a DB is excited, is under the influence of the periodic local potential induced by the surrounding atoms.

Figure 2b gives a schematic representation of an atomic chain in the field of a local periodic potential suppressing the thermal expansion of atoms. A hard nucleus of the Morse potential provides a larger contribution into the dynamics than its soft tail; the DB frequency increases beyond the phonon spectrum [26].

In a two-dimensional monoatomic Morse lattice one can excite a different DB with the radial symmetry (Fig. 3) [27]. Its frequency is also higher than that in the phonon spectrum because thermal expansion of its nucleus seems to be suppressed by the surrounding part of the crystal. The calculations demonstrate that in a two-dimensional case the DB nucleus causes radial displacements R , which decrease with distance from the DB center rather slowly, as R^{-1} , while the stresses decrease as R^{-2} [26, 27]. The radial compressive stress inhibits thermal expansion of the DB, which makes the contribution from the hard nucleus of the Morse potential sufficient for a DB with a hard type of nonlinearity to exist. In a three-dimensional case, the radial displacements decrease as R^{-2} , and the stresses – as R^{-3} [26, 27]. Therefore, suppression of thermal expansion of the DB by the surrounding elastic medium in the three-dimensional case is more efficient than that in the two-dimensional case. Therefore, a probability of formation and persistence of a discrete breather in a low-stability state of a three-dimensional system considerably increases compared to the two-dimensional case, and the more so compared to the one-dimensional case. A conclusion can be drawn that in two- and three-dimensional systems with interatomic potentials having hard nuclei and soft tails there can be DBs with a hard type of

nonlinearity, since the thermal expansion of the DB nucleus is suppressed by the surrounding part of the crystal, and the hard nucleus of the interatomic potential makes a larger contribution into the DB than does its soft tail.

DISCRETE BREATHERS IN PURE METALS

It was observed for the first time that DBs had rod-like structure (Fig. 2a) [28]; they could move along the close-packed atomic row. It was demonstrated that in pure metals there can be localized vibrational modes with a hard type of nonlinearity. This result was accounted for by the fact that in metals a contribution from odd anharmonicities is strongly reduced due to screening of the ion-ion interaction by the conduction electrons [29]. This is an additional source of increasing the bonding rigidity. Even the Morse pair potential admits an existence of a DB with a hard type of nonlinearity, in the case where its nucleus makes larger contribution into its dynamics than does the soft tail.

Initial conditions for exciting a DB. In certain crystals DBs can demonstrate a very high degree of spatial localization, such as e.g., band DBs in graphane [30], ionic crystals of the NaCl family with a significant difference between atomic masses of anions and cations [31], where DBs are localized practically on one light atom. In an extended grapheme, a DB is localized on a pair of atoms oscillating in antiphase [32–34]. Such DBs are readily excited by, e.g., setting an initial displacement for one atom or a pair of atoms having large oscillation amplitudes. In metals DBs are less localized (their structure is close to that shown in Figs. 2a and 3). Note that excitation of such collective vibrational modes is not a trivial task. In particular, under excitation of a rod-like DB in Nb, the initial displacements were set for eight atoms in a close-packed atomic row [28], and for excitation of a moving DB asymmetry was introduced into the initial displacements.

One of the approaches to searching the initial conditions for exciting a DB consists in analyzing delocalized nonlinear vibrational modes (DNVMs) [35]. The DNVMs are large-amplitude modes, which represent exact solutions of nonlinear dynamic equations and are subject to the point symmetry of the crystal lattice. If the frequency of DNVM leaves the phonon spectrum with the increasing amplitude, then to form a DB one can use a bell-shaped localizing function. This approach is efficient in exciting a DB in a two-dimensional Morse crystal [36].

In order to excite standing and moving rod-like DBs in metals, an ansatz (a physically grounded supposition in the form of a solution) was proposed with a number of parameters [26], which was successfully used for excitation of rod-like DBs in pure metals [37–39]. Such DBs can move along a close-packed atomic row at a velocity of about 0.1 of the velocity of light [38].

DB and crystal lattice defects. It is well known that crystal lattice defects to a large extent control the mechanical and physical properties of materials. The pioneering studies of a DB – vacancy interaction were performed within the frame of the Frenkel–Kontorova one-dimensional model [40]. It was shown that a moving DB, while interacting with a vacancy, can cause its migration. Furthermore, a significant role in this process belongs to the nonlinear vibrational mode localized on the atom neighboring the vacancy [40]. The migration of an interstitial atom due to its interaction with a moving DB was also studied. More recently, collision of a rod-like DB with a vacancy was studied in a two-dimensional Morse crystal. It was shown that a DB decreases the energy barrier of the vacancy migration [41].

The effects of X-rays or slow neutrons on a number of metals (Ni, Nb and Fe) were studied in [29]. It has been shown that an irradiation energy exceeding tens of electron volts gives rise to the formation of vacancies and interstitial atoms; note that a strong dependence on the direction of the irradiation pulse is observed.

The interaction of the moving rod-like DBs with a vacancy, a dislocation and a free surface in Fe was reported in [42]. In all cases, DBs decrease the energy barriers of defect migration, favoring the defect structure transformation in the crystal lattice.

DISCRETE BREATHERS IN ORDERED ALLOYS

Before a possible existence of DBs with a hard type of nonlinearity was studied in pure metals, band DBs with a soft type of nonlinearity was actively studied in the ordered alloys with large differences in the atomic masses of the

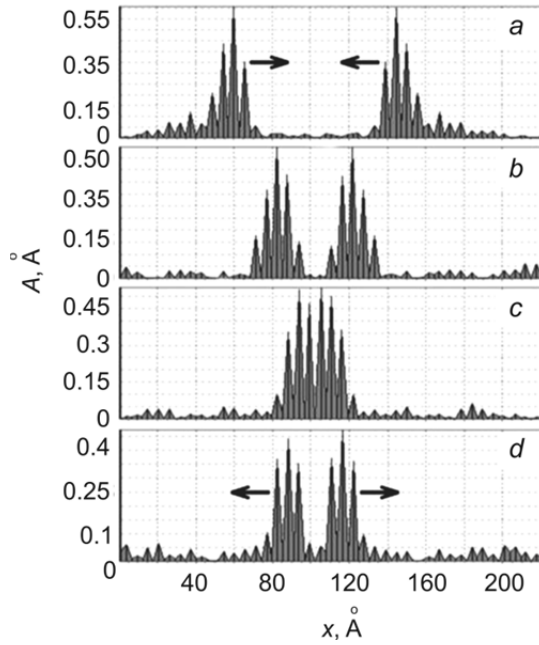


Fig. 4

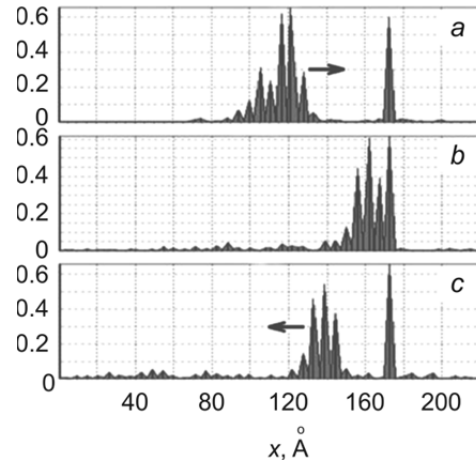


Fig. 5

Fig. 4. Collision of moving DBs in Pt_3Al . DBs interact in a nearly elastic manner repelling from each other with an insignificant energy lost on the emission of low-amplitude waves [44].

Fig. 5. Collision of a moving DB with an immobile DB in Pt_3Al . As a result of this collision the moving DB loses part of its energy on radiation and transfer to the immobile DB [44].

components. An intermetallic compound of Pt_3Al based on an FCC-lattice with the superstructure $L1_2$ was given a most detailed attention [43, 44]. One of the four sublattices of the translational cell contains the atoms of the same kind, and the remaining three – the atoms of the other kind. A wide gap in the phonon spectrum of the alloy is formed due to a large difference in the atomic masses of the components: $M_{\text{Pt}} = 195.23$ amu and $M_{\text{Al}} = 26.97$ amu. In [44], two types of DBs with the frequencies lying in the phonon spectrum gap and one type with the frequency lying higher than the phonon spectrum are analyzed.

An analysis of the collisions between the moving DBs (Fig. 4) demonstrated that they interact with each other in a nearly elastic fashion, repelling from each other. When a moving DB collides with an immobile DB [44] (Fig. 5), part of its energy is emitted, and is transferred to the immobile DB. Therefore the amplitude of the immobile DB after this collision appears to be somewhat larger than before the collision. The results [44] show that in the ordered alloys with a gap in the phonon spectrum it is possible to excite a large number of different types of spatially localized nonlinear vibrational modes.

An analysis of a possible excitation of DBs with a soft type of nonlinearity under a high-intensity external action in a Pt_3Al crystal [45] has revealed two possible mechanisms of exciting such breathers. In the first case, the excitation occurs after a single collision of the particle with the aluminum atom, and in the second case – by the energy accumulation on aluminum atoms in a series of successive collisions.

EFFECT OF DISCRETE BREATHERS ON PROPERTIES OF CRYSTALS

Many metallic materials function in strongly nonequilibrium and low-stability structural-phase states under the conditions of high-intensity external influences – irradiation, loading, and high-density electric currents. The materials

in these conditions are found in pre-transitional states characterized by low stability of their structure and properties towards external impacts [1–22]. One of possible manifestations of low stability consists in the spatially inhomogeneous distribution of the energy and its localization. It is in the region of pre-transitional low-stability states near the stability threshold that we can expect new scientific advances, effects and phenomena in the physical systems. For the region near the stability loss there are no validated physical concepts, models and approximations, and no theoretical approaches have been developed so far.

In this region, the role of structural defects (both static and dynamic) essentially increases [1–22]. The intrinsic structure inhomogeneity results in an inhomogeneous spatial distribution of the energy, primarily in local inhomogeneities in the form of lattice vibrations [1–22].

Given a low stability of the system towards excitations, the role of such localized crystal lattice vibration significantly increases. In essence these dynamic inhomogeneities are the precursors of structural-phase transformations in the system, with a variety of pathways of the transition depending on the type of structural inhomogeneity and the low-stability state of the system.

It is important to study the properties of DB due to their influence on the macroscopic characteristics. By now, a large body of knowledge about DB properties has been accumulated, which allows getting an insight into their role in the low-stability state of the systems near the stability threshold. Discrete breathers in metals are known to localize the energy up to about 10 eV [28, 29, 38, 39, 41–54], they can move along the close-packed rows of atoms at a velocity of ~ 0.1 of the sound velocity [39], transferring the energy to considerable distances. Discrete breathers can form due to thermal fluctuations and point structural defects (vacancies and interstitial atoms). The difference consists only in the fact that the formation of a structural defect gives rise to a certain energy increase, while DBs can form at a low energy, which can subsequently increase due to various pumping mechanisms [44].

The energy of a DB can be spent on the formation or migration of point and other defects [41, 42], on evaporation of atoms from the crystal surface [46], etc. They are the centers of phonon scattering and thus they decrease the crystal conductivity, contribute to the thermal capacity and thermal expansion of solids, and favor the generation of the second harmonic [55].

It is notable that the DB concentration can increase by the orders of magnitude under the conditions far from the thermal equilibrium, at considerably large energy flows through the crystal lattice (irradiation, plastic deformation, phase transitions, intense heating, shock wave propagation, etc.). This is due to the formation of localized low-stability states accompanied by structural transformations in the ordering alloy under thermal cycling [1–22] or in the study of acoustic emission during hot deformation of aluminum [6].

CONCLUSIONS

A large number of metallic systems function in strongly nonequilibrium and low-stability structural-phase states. A possible demonstration of low stability is an inhomogeneous spatial distribution of the energy and its localization. It is in the region of pre-transitional low-stability states that we can expect new breakthroughs in science. However, for the regions near the stability threshold there are no validated physical concepts, no models and approximations or developed theoretical approaches. The role of structural defects (static and dynamic) in these regions considerably increases. Given a low stability of the system towards excitations, the role of such localized crystal lattice vibrations becomes ever more important. In essence these dynamic inhomogeneities represent the precursors of the structural-phase transitions in the system.

A study of the properties of DBs is critical due to their influence on the macroscopic properties. By now, a sufficient body of knowledge has been accumulated to understand their role in the low-stability state of the systems.

It is noteworthy that interatomic interactions exhibit a hard type of nonlinearity at small distances and soft type – at large distances. Due to the asymmetry of nonlinear potentials at large vibrational amplitudes, the average interatomic spacing tends to increase. However, in two- and three-dimensional systems thermal expansion of the DB nucleus makes a larger contribution into the DB dynamics than does the soft tail. As a result, the DB frequency can increase with the amplitude and become higher than the phonon spectrum frequency. This suggests that the theoretical result (prohibition of existence of DBs with a hard type of nonlinearity) is valid for one-dimensional case only.

Even a simple two-dimensional lattice with the interatomic interaction in the form of the Morse potential can maintain at least two types of DBs, and three-dimensional systems can maintain at least three DB types. In the ordered alloys with a gap in the phonon spectrum a larger variety of possible DBs is expected.

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