===== **METALS** =====

Excitation of Gap Discrete Breathers in an A₃B Crystal with a Flux of Particles

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Abstract—The generation of discrete breathers in an A₃B crystal has been modeled by the method of molecular dynamics using Pt₃Al as an example via the application of random unidirectional momenta, which simulate the action of a particle flux, to atoms. Two possible mechanisms of the excitation of gap discrete breathers with a soft type of nonlinearity have been revealed depending on the energy of particles in a flux. If a particle is able to transfer energy of more than 1.4 eV to the Al atom, a discrete breather can be excited by the only particle. Otherwise, a discrete breather is formed upon numerous particle—Al atom collisions, which are possible only at a sufficiently high density of particles, as each following particle must transfer its momentum to the Al atom before its oscillations provoked by previous particles attenuate.

1. INTRODUCTION

Intense external actions on crystals (plastic strain, ultrasonic treatment, radiation, high-density electrical current, ion-plasma surface treatment, etc.) lead to considerable deviations of atoms from their lattice positions, activating different processes, which cannot be studied within the framework of linearized motion equations. Fluxes of particles (neutrons, electrons, etc.) may lead to heating and the formation of Frenkel pairs, pores, and other defects in crystals. Conversely, in polycrystals containing a great number of defects, they can result in defect annihilation and structural relaxation due to both material heating activating diffusion processes and direct interaction between a particle flux and defects. There currently exist many experimental works, in which it is pointed out that the observed phenomena cannot be explained by material heating alone and, consequently, this raises the problem of describing the athermal mechanisms of interaction between a particle flux and a crystal lattice and its defects. Let us cite only several examples. The ion-plasma surface treatment of high-purity germanium single crystals has led to the annealing of defects at a depth of several microns [1, 2]. Plasma treatment is confined to the bombardment of a crystal surface by ions with an energy ranged from 2 to 8 eV. It is interesting that mere heating does not lead to a similar effect of annealing for defects in germanium [1, 2]. In

the work [3], a magnesium alloy deformed by rolling was subjected to electrical discharge treatment. The material heating in the process of electrical discharge treatment was measured and calculated. It has been shown that pulse current promotes static recrystallization in the material, whereas mere heating gives no similar effect. The effect of thermal and athermal processes on the structure and properties of a functionally-graded titanium alloy subjected to electrical discharge treatment is discussed in [4]. The authors come to the conclusion that the phase and structural transformations observed in the alloy cannot be explained by the liberation of Joule heat alone. Electrical current passed through the metal in the course of its plastic deformation promotes an increase in plasticity and a decrease in yield stress [5–7]. Electrical current can be applied simultaneously with ultrasonic treatment of metals, thus improving the properties of their surface [8]. A decrease in yield stress due to irradiation by an electron beam with an energy of 0.5 MeV was also observed in polycrystalline aluminum and copper samples under their tension [9]. The migration of atoms due to high-density electrical current may change the morphology of the interface between two conductors [10–13]. The above described effects are most often explained with the use of phenomenological approaches, which give no insight into the atomistic mechanisms of interaction between a particle flux and a crystal lattice [3, 10–13]. The possibility to

excite spatially localized high-amplitude oscillatory modes called discrete breathers [14−16] with a particle flux is shown in this work. Discrete breathers localize energy up to several electronvolts and may be both mobile and immobile, and their lifetime ranges from several dozen to several thousand atomic oscillation periods, which may be sufficient to modify a defect crystal structure [16]. It is necessary to make a terminological reservation. In mathematical physics, discrete breathers are understood to mean strictly periodic oscillatory modes, which do not attenuate with time [14, 15], however, in real systems, where various disturbances are unavoidable, it is necessary to consider quasi-breathers characterized by nonstrict periodicity and a finite lifetime [17]. The quasi-breathers considered in this work will be called discrete breathers for short.

It is noteworthy that the existence of discrete breathers in alpha-uranium [18, 19] and an alkali halide NaI crystal [20, 21] was shown by experiment, and the active discussion of these results is continued [22, 23]. Molecular dynamic calculations confirm the possibility to excite discrete breathers in pure metals [24−30] and ordered alloys [31−40]. The first works on studying the properties of discrete breathers in low-dimensional crystals on the basis of first-principle calculations taking into account the electron structure of a substance have appeared [41−43].

According to recent works, discrete breathers can lead to the radiation-enhanced growth of pores in metals and the annealing of defects, make a contribution to diffusion, transport an electrical charge, decrease the energy barrier of chemical reactions in crystalline solids, etc. [1, 2, 26, 44−49].

Discrete breathers have a long lifetime, as their frequency lies outside the phonon spectrum of a crystal due to a nonlinear dependence between the frequency and the oscillation amplitude. The frequency of discrete breathers can decrease (increase) with growing amplitude, and such discrete breathers demonstrate a soft (hard) type of nonlinearity. Discrete breathers with a soft type of nonlinearity can exist only in crystals, whose phonon spectrum has a bandgap (gap), and their frequency lies within a spectrum bandgap, so they are called gap discrete breathers. Gap discrete breathers can be excited in biatomic crystals with a rather great difference between the atomic masses of their components [31−40, 50−53] and also in graphane [42, 43, 54] and uniformly deformed graphene [41, 55−57].

The objective of this work is to establish the possibility to excite a gap discrete breather with a soft type of nonlinearity in an L1$_2$-superstructure A$_3$B crystal with a particle flux. The selection of an A$_3$B-type alloy for studies is caused by a number of factors. L1$_2$-superstructure alloys with A$_3$B stoichiometry are numerous, and the search for the currently known L1$_2$-superstructure systems has allowed us to reveal nearly 190

such phases in the A$_3$B composition region. L1$_2$-superstructre alloys provide a basis for the development of currently existing superalloys [58]. Pt$_3$Al is considered as a model alloy. Materials of platinum group are of great interest due to high melting temperature and strength and exclusive stability under ambient conditions [59−61].

2. MODEL AND METHOD OF EXPERIMENT

The method of molecular dynamics was used to study the motion of atoms in ordered Pt$_3$Al alloy with an L1$_2$ superstructure on the basis of a FCC lattice (Fig. 1). A cubic translational crystal unit cell contains three platinum and one aluminum atoms. A computational unit cell comprising 16 × 16 × 16 of translational unit cells, i.e., 16 384 atoms, is considered. Periodic boundary conditions are used. Test calculations have shown that an increase in the size of the computational unit cell does not produce any effect on the obtained results, as dynamic objects with a high degree of spatial localization were used in this work.

The atoms interacted with each other via the pair Morse potential

$$\phi_{PQ}(r_{ij}) = D_{PQ}\beta_{PQ}\exp(-\alpha_{PQ}r_{ij}) \times (\beta_{PQ}\exp(-\alpha_{PQ}r_{ij}) - 1), \quad (1)$$

where D, β, and α are the potential parameters, and r_{ij} is the distance between atoms i and j. For Pt$_3$Al
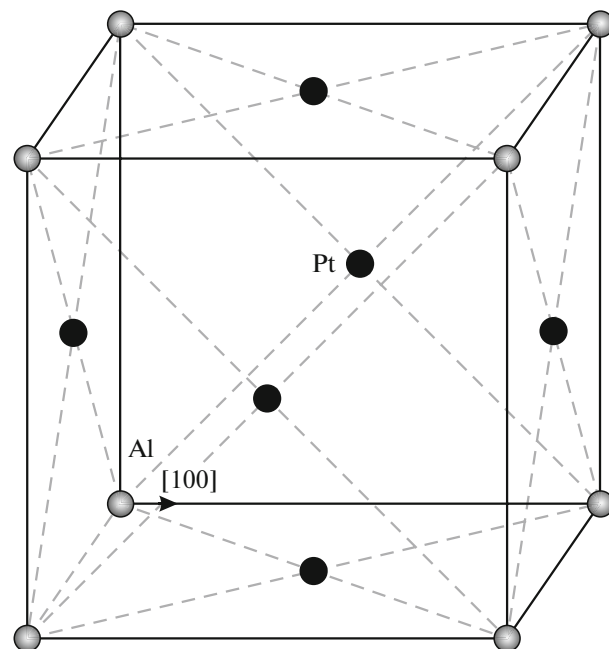
Fig. 1. Unit cell of ordered Pt$_3$Al alloy based on a FCC lattice. Al (Pt) atoms are shown in grey (black). Particle flux is assumed to move along crystallographic direction [100] (marked with an arrow), transferring momentum to lattice atoms.

alloy, the potential parameters were taken from the work [36]: $D_{AlAl} = 0.318$ eV, $\beta_{AlAl} = 27.4979$, $\alpha_{AlAl} = 1.02658$ Å^{-1}, $D_{PtPt} = 0.710$ eV, $\beta_{PtPt} = 102.89$, $\alpha_{PtPt} = 1.582$ Å^{-1}, $D_{PtAl} = 0.5048$ eV, $\beta_{PtAl} = 63.124$, and $\alpha_{PtAl} = 1.3501$ Å^{-1}. The procedure of fitting the potential parameters is described in the same work.

The transfer of energy from a particle flux to lattice atoms is modeled by increasing the momenta of a certain fraction of randomly selected atoms C in direction [100] by a specified value in such a fashion that the kinetic energy of each selected atom gains an instantaneous increment ΔE. Such a procedure is periodically executed with a time interval T. The range of energy transferred to each randomly selected atom was varied from 0.2 to 5 eV per interaction. This energy was received by a fraction of computational unit cell atoms within a range from $C = 0.00025$ to $C = 0.015$. The periodicity of momenta T was varied within a range from 0.5 to 10 ps. These parameters were selected such that no topological defects (Frenkel pairs, etc.) appeared in a crystal.

A gap discrete breather with a soft type of nonlinearity in a Pt$_3$Al crystal represent one Al atom oscillating in direction [100] at a great amplitude, whereas its neighboring atoms have much smaller oscillation amplitudes [34]. It has been established that the minimum energy of a gap discrete breather in a Pt$_3$Al crystal amounts to nearly 0.8 eV, but the excitation of a discrete breather requires slightly more energy, as the energy transferred to an individual Al atom is partially dissipated over the lattice upon the formation of a collective oscillatory mode. Hence, an Al atom must accumulate the required energy at the expense of one or several received sequential momenta.

The energy ΔE transferred to a lattice atom can be calculated from the momentum and energy conservation laws, assuming that a collision is perfectly elastic. ΔE will be maximal upon a central collision, and we have for nonrelativistic particles that

$$\Delta E = 4M_1 M_2 E_0 / (M_1 + M_2)^2, \qquad (2)$$

where M_1 and M_2 are the masses of an incident particle and a lattice atom, respectively, E_0 is the energy of a particle, and a lattice atom is assumed to be immobile before collision.

3. MODELING RESULTS

Calculations have shown that a discrete breather can appear in the case of particles with a rather high energy at the expense of the only momentum transferred to an Al atom. The generation of a discrete breather in a Pt$_3$Al crystal is exemplified in Fig. 2 for $\Delta E = 5$ eV. According to Eq. (2), this energy can be received by an aluminum atom upon collision, e.g., with a very slow neutron, whose energy E_0 is nearly 35 eV. Let us note that momentum was transferred to
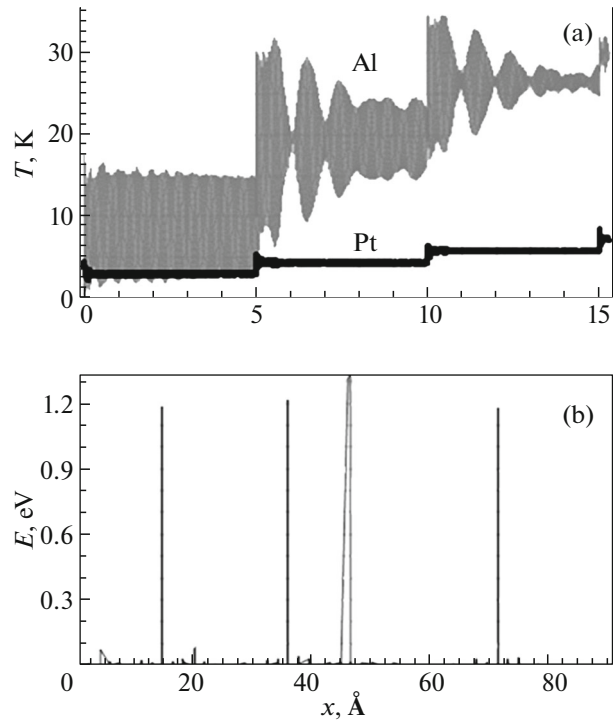
Fig. 2. (a) temperatures of the Pt (black) and Al (grey) sublattices versus time for the interaction with a particle flux with an energy $\Delta E = 5$ eV. Momentum was transferred from a particle flux to the lattice every $T = 5$ ps. Every time, momentum was received by 8 randomly selected atoms; (b) distribution of energy in a crystal along the X axis at a time moment of 9 ps.

the fraction of computational unit cell atoms $C = 5 \times 10^{-4}$ every $T = 5$ ps. Taking into account the total number of atoms in the system, we find that momentum was received every time by 8 randomly selected atoms. In Fig. 2a, the Pt and Al sublattice temperatures (black and grey lines, respectively) are plotted as functions of time. The distribution of energy in a crystal along the X axis at a time moment of 9 ps is illustrated in Fig. 2b. By this time moment, the crystal has received two portions of momenta at a total number of excited atoms of 16. Statistically, according to the crystal stoichiometry, nearly 1/4 of them are light Al atoms able to create a discrete breather. As can be seen from Fig. 2b, 5 discrete breathers (two of them at $x \approx 46$ Å) in a crystal has turned out to be excited by the time moment $t = 9$ ps, thus being in agreement with the expectation.
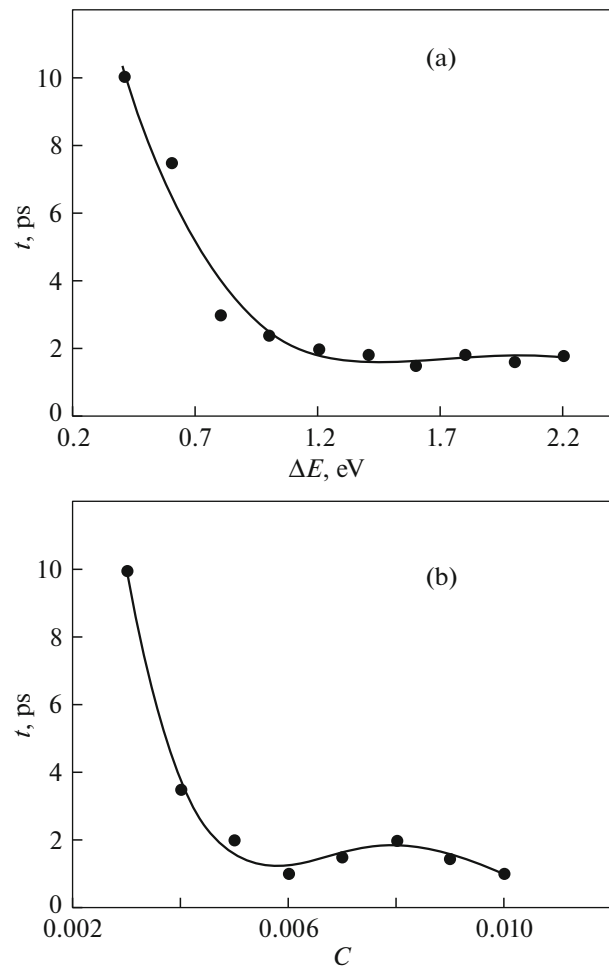
An example for a particle flux of lower energy, when a single transfer of momentum to an Al atom is insufficient to excite a discrete breather in it is illustrated in Fig. 3. In this case, $\Delta E = 1$ eV, $T = 0.5$ ps, and $C = 0.005$. By the time moment $t = 5$ ps, a crystal has received nine momenta transferred to 819 atoms, and it is evident that a certain fraction of these atoms received momentum several times. As a result, four discrete breathers, namely, two with an energy pf
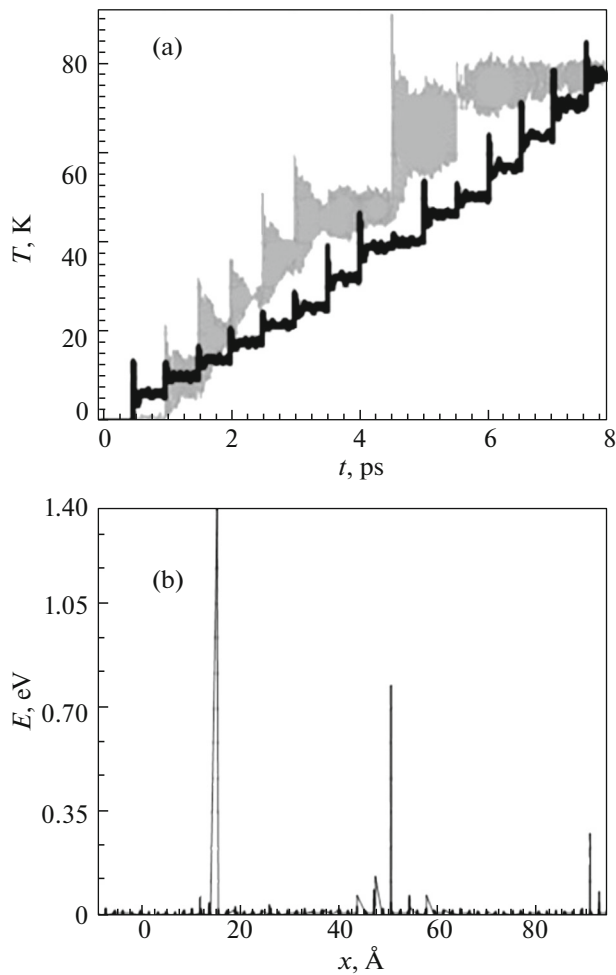
Fig. 3. (a) temperatures of the Pt (black) and Al (grey) sublattices versus time for the interaction with a particle flux with an energy $\Delta E = 1$ eV at $C = 0.005$. Momentum was transferred from a particle flux to the lattice every $T = 0.5$ ps. Every time, momentum was received by 8 randomly selected atoms; (b) distribution of energy in a crystal along the X axis at a time moment of 5 ps.

Fig. 4. Time of first discrete breather appearance in a computational unit cell versus (a) energy transferred by particles ΔE at fixed $T = 0.5$ ps and $C = 0.005$ and (b) C at fixed $\Delta E = 1$ eV and $T = 0.5$ ps.

1.4 eV, one with an energy of 0.8 eV, and one breather attenuating by the moment of recording the results with an energy of nearly 0.3 eV, has turned out to be excited in a crystal.

Hence, it is possible to reveal two methods of exciting discrete breathers with a soft type of nonlinearity in a Pt₃Al crystal with a particle flux. When particles transfer a rather high energy, each collision with an Al atom leads to the excitation of a discrete breather. When the energy transferred from a single particle is insufficient to excite a discrete breather, it can be excited upon sequential excitation by more than one particles, if they collide with the atom in a rather short time interval, when its oscillations appearing upon collision with previous particles have not enough time to attenuate. Such pumping of a discrete breather with

energy is possible on condition that a low-energy particle flux is sufficiently dense.

The excitation time of the first discrete breather in a crystal was found at different parameters of a particle flux. The results for the case of a varied energy ΔE of particles in a flux are shown in Fig. 4a. Here, $T = 0.5$ ps and $C = 0.005$ are fixed. It can be seen that discrete breathers can appear at $\Delta E > 1.4$ eV upon interaction of an Al atom with a single particle, and the transfer of momentum to an Al atom from a greater number of particles is required at lower energies, and each following particle must transfer its momentum no later than the Al atom oscillations produced by the interaction with previous particles attenuate. At energies $\Delta E < 0.4$ eV, no discrete breathers were observed in the computational unit cell of selected size. This is due to the fact that an Al atom must receive several sequential momenta for a short time period at such a low energy of particles to excite a discrete breather, but

the probability of such an event for the selected parameters is small.

In Fig. 4b, the excitation time of the first discrete breather in a crystal is plotted as a function of C for the case of $\Delta E = 1$ eV and $T = 0.5$ ps. At $C > 0.005$, the time of waiting for the first discrete breather is small and weakly depends on C. At lower C, the time of waiting for the first discrete breather growth with decreasing C due to the fact that the sequential excitation of an Al atom in a short time period before its oscillations produced by previous momenta attenuate is of low probability in the case of an insufficiently dense flux of particles.

It is noteworthy that the lifetime of discrete breathers generated by a particle flux is short, and they exist for 7−15 oscillation periods, which correspond to 0.56−1.12 ps. The lifetime of discrete breathers is reduced due to an increase in the temperature of a unit cell with each following momentum. Nevertheless, high-amplitude atomic oscillations excited by thermal fluctuations in crystals, which do not maintain the existence of discrete breathers, last no longer than a single oscillation. Hence, the lifetime of discrete breathers is an order of magnitude greater than for high-amplitude thermofluctuation oscillations of atoms.

4. CONCLUSIONS

The computer experiments performed by the method of molecular dynamics have shown that a particle flux is able to excite gap discrete breathers with a soft type of nonlinearity on Al atoms in a Pt_3Al crystal with a lifetime of nearly ten atomic oscillations, which is an order of magnitude greater than the lifetime of high-amplitude thermal fluctuations in crystals, which do not maintain the existence of discrete breathers. Two mechanisms of the excitation of discrete breathers depending on the energy of particles in a flux have been revealed. When particles are able to transfer energy of more than 1.4 eV to Al atoms, discrete breathers can be excited by the only particle. When the energy of particles is lower than 1.4 eV, discrete breathers are formed upon numerous collisions of particles with Al atoms, but this is possible only at a rather high density of particles, as each following particle must transfer its momentum before the Al atom oscillation produced by previous particles attenuate. A growth in the energy of particles and the density of a particle flux increases the probability of the excitation of discrete breathers.

The fact established in this work that a particle flux is able to excite discrete breathers in crystals opens new opportunities for the interpretation of the experimental data [1−13] indicating a considerable contribution of athermal effects to the dissipation of particle flux energy. Discrete breathers, which are not topological defects, but accumulate the energy of nearly 1 eV and are able to exchange by this energy [16], may influence a defect structure of crystals for the time of their existence [1, 2, 16, 46−48]. Hence, the energy of discrete breathers will be eventually converted into heat only in part, and its other portion will be spent on the structural transformation of a crystal.

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