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THE EFFECT OF THERMAL VIBRATIONS ON THE ACCELERATION OF NEUTRAL ATOMS DURING A SURFACE PHASE TRANSITION

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The paper explores the effects of thermal vibrations on the acceleration of atoms during a surface phase transition which occurs via the coherent energy transfer mechanism from many adatoms initially in metastable states to a single accelerated adatom. The acceleration is the result of a long chain of subsequent collisions with a single particle harvesting the energy of the transition of its predecessor in the chain. The paper presents the analysis of the stability of the suggested system, simulation of the dynamics of the accelerations and the analysis of the limits imposed on the process by defocusing and scattering on thermal vibrations.
Key words: acceleration, surface, phase transition.

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I. INTRODUCTION

A series of publications [1–3] suggested a mechanism of the acceleration of neutral atoms in a non-equilibrium system during its transition to the equilibrium state. The neutrality of the accelerated atoms makes the proposed mechanism distinctly different from the conventional ways of particle acceleration [4, 5]. A prerequisite for the acceleration is the existence of two energy minima for atomic positions along a certain direction in the crystal. The acceleration occurs when the atoms initially occupy high energy positions. Qualitatively, the considered acceleration effect can be presented as follows. Let us imagine a system in which in a certain direction there are two potential minima for atoms and, initially, the atoms are situated in the upper minima. Such a state is metastable and with time the atoms would move to the low energy positions. They can relax separately. However, as the papers [1–3] show, the relaxation can occur in a coherent way, the atoms along an atomic chain would undergo subsequent transitions transferring their energy to the next one in the row. Let one of the atoms acquire a certain energy (from an external source or fluctuationally), which is sufficient for overcoming the potential barrier, and collide with the neighbor atom. As is known after a central collision of a moving and a stationary balls of equal mass the moving ball stops and the stationary ball starts to move. If after the collision the first atom stops in the vicinity of the lower minimum (point A in Fig. 1) then it transfers the energy released at the relaxation to the second atom. As a result, the second atom accelerates. These accelerations are accumulated and, eventually, the energy passed to the last atom in the collision chain can be very large, provided the number of subsequent collisions is sufficient.

The focusing phenomenon known in radiation physics facilitates the acceleration. The focusing confines the motion of atoms to a certain crystal direction, if their energy does not exceed a certain threshold value in the range of hundreds of eV.

Several systems where such an acceleration may occur were considered in [1–3, 6]. The acceleration is possible during structural phase transitions in strongly strained crystals, at a movement of a screw dislocation, in a system with a "domino"-type photo-induced phase transition [7]. The term "acceleron" has been coined in papers [1–3] to refer to the process in a crystal when a sequence of atomic collisions accelerate a single atom. The estimates of different mechanisms of the energy loss at collisions [1–3] show that the energy of an acceleron may be as high as a hundred electron-volts. As the energy of the accelerated particle is much greater than kT, accelerons could be one of the reasons for a number of anomalous phenomena: the emission of the electromagnetic waves in the UV and X-ray spectral ranges as a result of atomic excitations at collisions, the increase in the probability of the low-threshold nuclear reactions, etc.

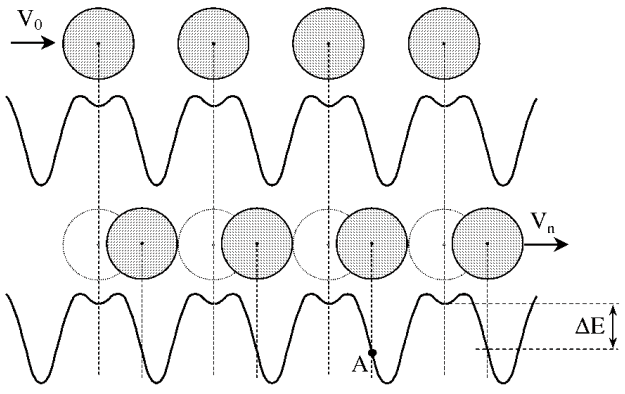

Fig. 1. Schematic illustration of the acceleration mechanism. At the top the initial metastable state is shown. The bottom row shows the process during which each of the atoms in the chain is dispaced prior to a collision to a position where its potential energy is smaller than the energy in the initial state. Accordingly, the next atoms in the chain gains the same value of the kinetic energy.

This paper presents a simulation of the acceleration process that happens on the flat surface during a surface phase transition. As shown in [2] one of the systems where accelerons can appear is a crystal in a state preceding a phase transition where the inversion of the population of the crystal sites could arise at the boundary between different phases. There is a great variety of structural phase transitions in systems of adatoms on the crystal surface [8–10]. A model of one of such systems is studied in the present paper where the possibility of acceleration is confirmed and the effects limiting the acceleration are considered: defocusing and thermal vibrations that lead to the random non-central collisions.

II. MODEL

We consider a system of adatoms on a crystal surface assuming that they can form two different lattices comprising two phases, one of which is stable and the other is metastable. Our purpose is to study the dynamics of the phase transition during which an acceleron may appear.

In the proposed model, the adatoms above the crystals surface interact with each other and also are affected by a potential imposed by the surface. The total potential energy is, thus, given by an expression

$$U(\{\mathbf{r}\}) = \sum_i U_{\text{ex}}(\mathbf{r}_i) + \frac{1}{2}\sum_{i\neq j} U_{\text{in}}(\mathbf{r}_i - \mathbf{r}_j), \qquad (1)$$

where $\mathbf{r}_i$ are coordinates of the i-th atom, U_{in} is the potential of the inter-atomic interaction, U_{ex} is the external periodic potential, created by the atoms of the substrate.

We have chosen the Lennard-Jones potential for the interaction between the atoms:

$$U_{\text{in}} = U_0\left(-\frac{r_0^6}{r^6} + \frac{r_0^{12}}{2\,r^{12}}\right) \qquad (2)$$

Further on we use such dimensionless units that $U_0 = 1$ and $r_0 = 1$. The length units are such that the Lennard-Jones potential has a minimum at $r = 1$. For realistic systems the unit of energy may vary in the range of $0.5 \div 3\,\text{eV}$.

When the interaction with the surface is not taken into account, a two-dimensional system of adatoms interacting via a spherically symmetrical potential would form a hexagonal lattice. We assume that the surface imposes on adatoms an additional field in a manner to prevent this from happening. The external potential is chosen to define a square grid and takes the form

$$U_{\text{ex}}(x,y) = U_e\left(-\cos\frac{4\pi x}{a} - \cos\frac{4\pi y}{a} - \frac{1}{4}\cos\frac{8\pi x}{a} - \frac{1}{4}\cos\frac{8\pi y}{a}\right) \qquad (3)$$

The two last cosines in the potential do not create additional minima but help to make the wells narrower and the barriers smoother.

We shall consider a system in which the adatoms occupy a quarter of the potential wells defined by the grid (every second minimum in each direction). The coefficient U_e, which determines the interaction of adatoms with the surface is considered to be small, while at the same time strong enough to prevent adatoms from forming a hexagonal lattice.

Thus, when the atoms occupy every second site of the grid along each row one can construct two types of lattices shown in Fig. 2: a square lattice and a lattice with a shifted row which could be classified as a rectangular centered. We shall consider a phase transition between those two lattices. Each of them has the same energy in the external potential of the substrate, but their energies differ due to the inter-atomic interaction. In the lattice with shifted rows the distance between the atoms is larger but, at the same time, the number of the closest neighbors increases. The balance between those factors determines which of the lattices is more stable depending on the grid period a. Calculations show that the square lattice becomes more stable at $a > 1.05825$ in the units of r_0. For a smaller period of the external potential the lattice with the shifted rows is energetically preferable.

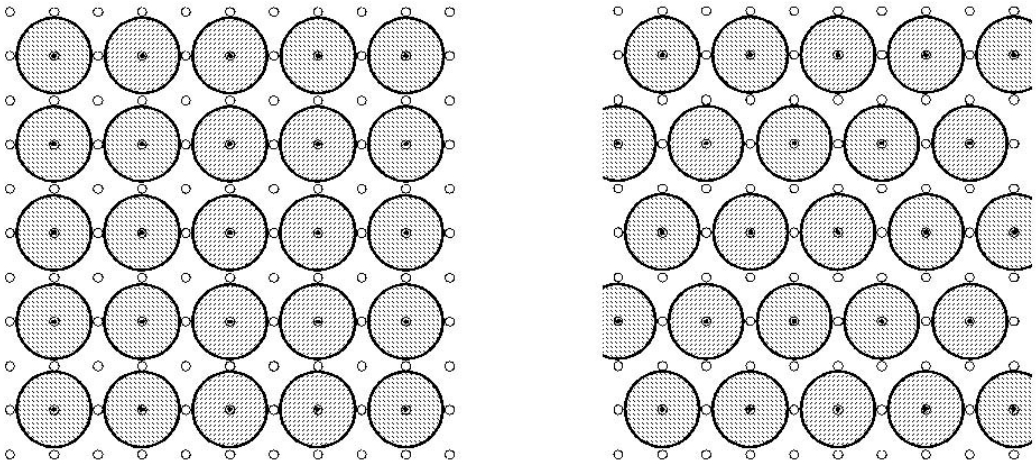

Fig. 2. Two types of lattices adatoms may form above the grid defined by the surface: a square lattice is on the left and the lattice with shifted rows is on the right.

Without stabilizing interaction with the substrate a two dimensional system of spherical atoms would form a hexagonal close-packed lattice with the symmetry different from both proposed arrangements. This fact is a source of a possible instability of the system.

We suggest studying mechanisms of the phase transition from the initial square lattice to the lattice with the shifted rows. We assume that the initial square lattice of adatoms has been formed, for example, at higher temperature when the lattice of the substrate is more spacious. At cooling or as a result of the hydrostatic compression the period of the substrate-imposed potential may decrease and the system of adatoms would find itself in a metastable state.

III. ANALYSIS OF STABILITY

We consider the case when the adatoms occupy the sites determined by the substrate potential. The stability of the arrangement of atoms is analyzed both for the square lattice and for the lattice with shifted rows. The criterion of the stability of the system is the condition that the matrix of the second derivatives of the potential with respect to the displacement of atoms from equilibrium position would be positively determined.

Coordinates of the i-th adatom may be written in the form $\mathbf{r}_i^p = \mathbf{r}_i^0 + \mathbf{r}_i$, where $\mathbf{r}_i^0$ are the coordinates of the lattice sites and $\mathbf{r}_i$ are a small displacement which can be presented in the form $\mathbf{r}_i = \sum_{\mathbf{k}} \mathbf{r}_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{r}_i^0)$

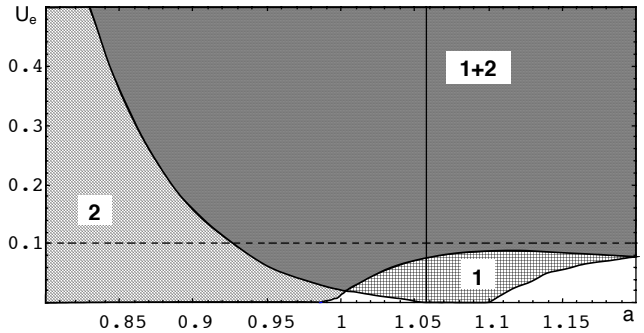

Fig. 3. Regions of stability of two types of lattices against the surface grid period a and the value of the surface potential U_e. The region of stability of the square lattice is denoted as 1, the same for the lattice with shifted rows is denoted as 2. In the 1+2-region of parameters both lattices are stable.

Expansion of the potential of Eq. (3) in series of displacements is obtained after some straightforward transformations

$$U(\{\mathbf{r}_i\}) = \sum_i U_{\rm ex}(\mathbf{r}_i^p) + \frac{1}{2}\sum_{i\neq j} U_{\rm in}(\mathbf{r}_i^p - \mathbf{r}_j^p) = U_0 \qquad (4)$$

$$+ N\sum_{\mathbf{k}} |x_{\mathbf{k}}|^2 \left(\frac{16\pi^2 U_e}{a^2} + \sum_{i\neq 0} \frac{\partial^2 U_{\rm in}(\mathbf{r}_i^0)}{\partial x^2} \sin^2 \frac{\mathbf{k}\mathbf{r}_i^0}{2} \right)$$

$$+ N\sum_{\mathbf{k}} |y_{\mathbf{k}}|^2 \left(\frac{16\pi^2 U_e}{a^2} + \sum_{i\neq 0} \frac{\partial^2 U_{\rm in}(\mathbf{r}_i^0)}{\partial y^2} \sin^2 \frac{\mathbf{k}\mathbf{r}_i^0}{2} \right)$$

The factors at $|x_{\mathbf{k}}|^2$ and $|y_{\mathbf{k}}|^2$ should be positive for all values of $\mathbf{k}$ if the atomic positions in the sites are minimums.

Fig. 3 shows the diagram of stability of the square lattice and of the lattice with the displaced rows depending on the period of the surface grid a (x-axis) and the amplitude of the external potential U_e (y-axis). To the left of the vertical line at $a = 1.05825$ the lattice with the shifted rows has smaller energy while to the right of that line the square lattice has smaller energy.

Numerical calculations show that even without the external potential the square lattice is locally stable at $1.06923 < a < 1.09512$. For example, at $U_e = 0.1$ in units of U_0, the square lattice is stable when $a > 0.92607$.

IV. METASTABLE ATOMIC POSITIONS

The stability analysis shows that in a certain region of parameters there exist two possible states for the lattice of adatoms, one of which is stable and the other is metastable. In the metastable state a row of atoms is sandwiched between its neighbors and has two local minima separated by the period of the grid (the half period of the lattice of adatoms), which correspond to the different lattices. A transition of a row from one minimum to another is a structural phase transition.

The mechanism of the acceleron creation is described qualitatively in Introduction. The energy gain in each subsequent collision depends on the point where the atom stops. If the atom stops close to the well bottom then the energy gain is large and when it stops close to the highest point of the potential barrier then the energy gain is small. The particles can even lose energy if they are stopped at the top of the potential barrier.

V. MODELING AND RESULTS

Molecular dynamic modeling was performed for the 250×8 strip of atoms initially located on the square grid with the period $a = 0.96$ and for the value of the amplitude of the external potential $U_e = 0.1$. We numerically solved the system of Newton equations:

$$m\frac{d^2\mathbf{r}_i}{dt^2} = -\nabla_i U(\{\mathbf{r}\}), \qquad (5)$$

where $U(\{\mathbf{r}\})$ is given by Eq. (1) and m is the mass of an atom. Our simulation software is using fourth order Runge–Kutta method with a time step of 1/150 in dimentionless time units.

As mentioned already, in order to start an acceleron, an atom has to overcome the potential barrier. This may happen either due to an impact of external origin or due

to thermal fluctuations. In this paper the former situation is considered. We studied the region of temperatures sufficiently low to make the cases of the thermal fluctuation caused transitions of atoms from the upper to the lower well extremely rare. Therefore, if the atoms are initially in the vicinity of the upper wells, then they will stay there indefinitely long. The temperature effect in the initial system is reduced to the equilibrium vibrations within the metastable state.

For the simulations all atoms were initially placed in their respective lattice sites and assigned small velocities, Gaussian distributed with a certain small mean energy, and the system was allowed to equilibrate for a certain period of time. At the start of the simulation we observed that the kinetic energy decreased by a half and the atoms in the grid acquired some displacements from the initial positions at the lattice sites. Later the situation stabilized. We believe that during this initial period the atomic motion acquired a temperature defined by the chosen mean energy and all correlations were properly set. After that we have manually assigned a significant velocity along the x-axis to one atom at a certain lattice site. By doing this we simulate the interaction of an atom with a high energy particle of a fluctuation that we cannot achieve by the simulation process. The value of the velocity was chosen sufficient to start the process of the acceleration.

The program tracked the atoms with the largest velocity and when their distance from the end of the strip was less than 150 lattice periods the modeled region moved forward by a period: new atoms were added at the end of the strip while the atoms at the beginning of the strip were removed. By using this procedure we were able to follow the acceleration for a significant period of time by running dynamics of a small number of atoms. As the acceleron is faster than the other oscillations in the system the dynamics of the atoms left behind does not affect its propagation while the added atoms were far enough to justify addition at the fixed position in the lattice sites. The number of atoms between the high-velocity atom and the boundary of the simulation window allowed us to expect that the thermal equilibrium is always set in the region of the acceleron prior to its arrival.

First we simulated the acceleration at practically zero temperature when the energy of atomic vibrations was 10^{-5} in dimensionless units. In Fig. 4 we show the time dependence of the velocity of every 50-th atom along the acceleron path. As the acceleron propagates, the velocity of each following atom increases, adsorbing in a coherent way the energy released at the relaxation. For faster atoms the time interval between the peaks decreases. One can see that the energy accumulates in the form of the kinetic energy of a single atom.

As the acceleron propagates along the crystal the atoms that are left behind are shifted by a period of the substrate grid. In this manner the lattice behind the acceleron changes its symmetry. During the time of simulation other rows were not affected by the transition and the remaining part of the released energy (which was not accumulated by the accelerated atom) dissipated.

The analysis of the time dependence of the positions of atoms that were left behind the acceleron shows that the faster they move the closer to the well bottom each atom stops. Therefore, the energy transferred further along the row is larger and the amplitude of the subsequent vibrations is smaller. The efficiency of the acceleration grows with each collision as illustrated by the plot in Fig. 6.

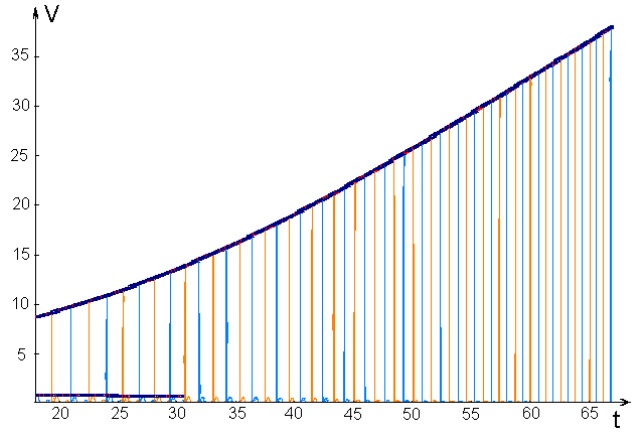

Fig. 4. Time dependence of the velocity of each 50-th atom in the collision chain in the simulation. The plot clearly shows gradual acceleration of the adatoms in the chain of collisions.

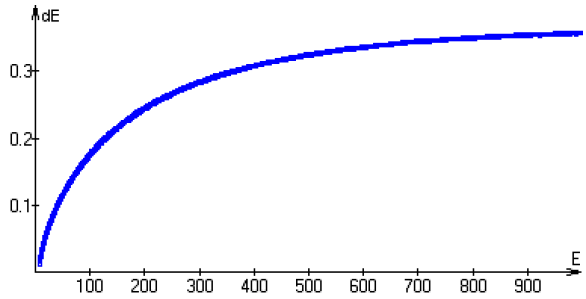

Fig. 5. Energy gain versus energy as calculated at zero temperature

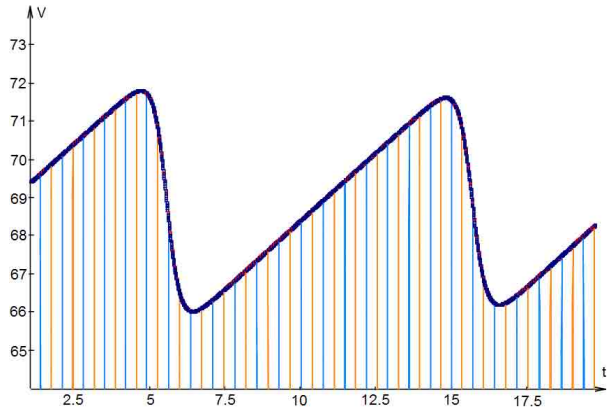

Fig. 6. The defocusing effect. The velocity of each 50-th atom in the collision chain is shown as a function of time. Defocusing leads to the decrease of the kinetic energy of the fastest atom. The acceleration process picks up again after the velocity decreases below the level of defocusing.

VI. DEFOCUSING

There are several factors limiting the energy of the accelerated atom: structural defects, impurities, thermal oscillations and defocusing. These effects have been considered in the papers [2, 3]. They can be decreased by a choice of a more perfect system and by lowering the temperature. Yet the defocusing cannot be suppressed or eliminated. We have carried out its modeling. The propagation of an acceleron requires availability of straight rows of atoms. The accelerating propagation occurs along this row. Moving at an angle to this direction an atom experiences a non-central collision. In this case its kinetic energy cannot be fully transferred to the next atom in the chain: a part of the energy stays with the incident atom and a part in dissipated to the vibrations in the perpendicular direction. The losses are larger for greater values of the deviation. Yet, at small energies there exists a known focusing effect: after each subsequent collision the deviation of the direction of the atomic motion from the line formed by the atomic row decreases (see [11]). According to the calculations the focusing effect holds up to the energy of several hundred eV, disappearing at larger energies. When the accelerated particles reach that value of energy a random deviations from the direction of propagation would tend to increase.

The results of the modeling of the defocusing effect are shown in Fig. 6. The simulation has been performed in the vicinity of the threshold velocity for the thermal vibration of the order of 10^{-3}. For the chosen parameters the threshold value of velocity in dimensionless units is 69. In order to reach that velocity the atoms in the row have to experience approximately 1000 collisions. After that the energy losses due to the defocusing effect prevail and the propagation of the accelerons slows down. Further simulations show that in this particular model system the acceleration starts anew after the energy of the fastest particle becomes sufficiently small.

VII. SCATTERING ON THERMAL VIBRATIONS

The increase of the amplitude of thermal vibrations affects the scenario of the events in such a way that the acceleration slows down. In order to evaluate this effect we performed simulations at different temperatures. The results are summarized in Fig. 7.

At non-zero temperatures one can see the decrease of the acceleration rate due to the energy dissipation on thermal vibrations. Moreover, occasional greater energy losses are seen, believed to be due to the dissipation on so big thermal fluctuations that the focusing needs about a dozen of collisions to reset one-dimensionality of the acceleron.

Each temperature sets a limit for the highest energy the atom may acquire. This is clearly seen from curve 2 in Fig. 7. If the initial energy exceeds this maximum the atoms slow down as seen from curve 4. As calculations show the limit energy does not depend on the initial energy of the starting atom. If the initial energy is larger than the limit the atoms are decelerated, if the initial energy is smaller than the limit energy the acceleration occurs.

Fig. 8 depicts the largest energy that the accelerated atom acquired as the function of temperature. At low temperatures, the limit energy of the accelerated atom is large. It drops fast with increasing temperature. At a higher temperature the acceleration gain becomes small due to the scattering on thermal vibrations.

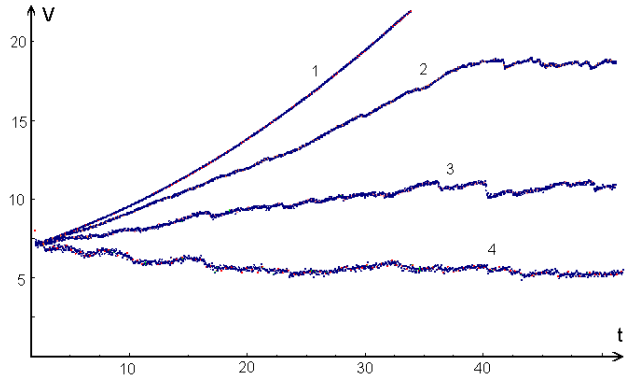

Fig. 7. The acceleration curves for simulations performed for different temperatures of the system. 1: $T = 10^{-5}$; 2: $T = 0.001$; 3: $T = 0.0066$; 4: $T = 0.01$.

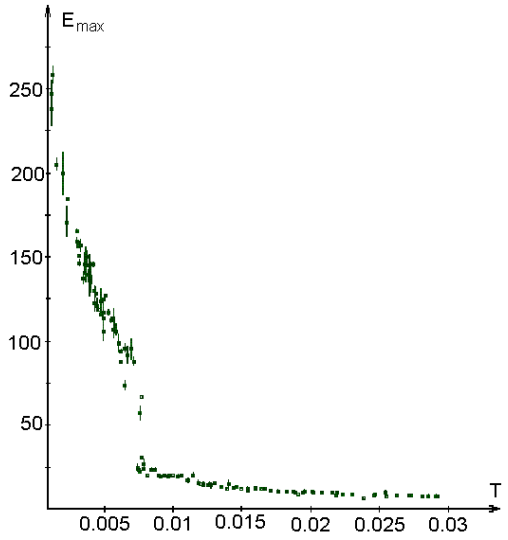

Fig. 8. The largest energy of the accelerated particle versus temperature.

All graphs are plotted in dimensionless units where the energy unit is defined by the value of the interatomic interaction. Let us assume for the estimates that the energy unit $U_0 = 1$ eV. Then, as Fig. 8 shows, the limit energy at the temperature of 150 K is about 30 eV.

The simulations revealed also a possibility of two-atom acceleration which happens when the energy of the atom is not sufficient to push it deep in the potential well. In this case the atom stops beyond the barrier yet with a large potential energy. This energy can be released in

the coherent way as well producing a second accelerated atom following the first. The second accelerated atom is faster, it reaches the first and gives it an additional push slowing itself, then gains the energy again and so on. As the result two moderately accelerated atoms may leave the system.

VIII. CONCLUSIONS

The analysis of the requirements to a system in which accelerons are possible has been performed. The limitations imposed on the acceleration by the temperature effects have been investigated. In the studied model of the acceleration one of the atoms on the initial grid was given certain energy in order to overcome the potential barrier between the atoms. In real conditions such energy may be communicated to an atom by the external low energy beam of particles or as the result of fluctuations. Acceleration in a fluctuating system as well as the energy distribution of the accelerated atoms require further studies that are planned following of the current research.

So far the accelerons were not observed experimentally. Yet no experiments that would intentionally look for them were performed. At the same time the fast moving atoms in crystals may manifest themselves in the appearance of the processes, the triggering of which requires energy significantly exceeding kT. Some anomalous phenomena in crystals have not found a satisfactory explanation while accelerons may play a role in them. A number of such phenomena has been analyzed in [12]. They include the visible optical emission arising at the ultrasonic irradiation of semiconductors, cold nuclear synthesis, anomalous mass transfer, etc.

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ВПЛИВ ТЕПЛОВИХ КОЛИВАНЬ НА ПРИСКОРЕННЯ НЕЙТРАЛЬНИХ АТОМІВ ПРИ ПОВЕРХНЕВОМУ ФАЗОВОМУ ПЕРЕХОДІ

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У цій роботі вивчено вплив теплових коливань на прискорення атомів при поверхневому фазовому переході, який здійснюється через механізм когерентного переносу енергії від багатьох адатомів у початкових метастабільних станах до одного адатома, що прискорюється. Прискорення є результатом довгого ланцюжка послідовних зіткнень із однією частинкою, що збирає енергію переходу своїх попередників у ланцюжку. У статті представлено аналіз стійкості запропонованої системи, симуляцію динаміки прискорень та аналіз обмежень, що накладаються на процес дефокусуванням та розсіянням на теплових коливаннях.