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Theoretical study of magnetic properties and multiple twin boundary motion in Heusler Ni-Mn-Ga shape memory alloys using first principles and Monte Carlo method

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Abstract. In this paper we propose a microscopic model for description a twin boundary motion in Ni_2MnGa Heusler alloy by means of the *ab initio* and Monte Carlo calculations. The tetragonal real lattice of martensite with $c/a < 1$ was used in our simulations. The variants of the low temperature martensite are separated by the twin boundary. The complex Hamiltonian consisting of the Heisenberg model and Blume-Emery-Griffiths one are considered. On the basis of the proposed Hamiltonian the influences of external magnetic field and anisotropy on the twin boundary motion are studied. It is shown that the theoretical picture of the twin boundary motion is analogous with experimental observations.

Introduction

Currently the ferromagnetic Heusler alloys have attracted a much attention in view of their unique properties such as shape memory effect, giant magnetocaloric effect, magnetostriction, superelasticity, motion of twinning martensitic variants [1]. As regards the last property the mechanism of deformation twinning by the external forces is interesting physical phenomenon suitable for device applications [2]. For example, in the martensitic state, the twin boundary motion due to the reorientation of variants can be induced reversibly by an external magnetic field. This effect is discovered in many Heusler alloys, for example in Ni-Mn-X (X= Ga, In, Sb, Sn) ones. In this paper we present the theoretical microscopic model for description of twin boundary motion in the stoichiometric Ni_2MnGa compound.

Theoretical model

In the proposed model, we consider a three-dimensional real tetragonal lattice ($c/a < 1$) of Heusler alloys with periodic boundary conditions. We take into account the exchange nearest-neighbor interactions in the first coordination shell of each atom only. The magnetic exchange interactions between the Ni and Mn atoms in Ni_2MnGa alloy have been taken from our previous *ab initio* simulations using the spin-polarized relativistic Korringa-Kohn-Rostoker (SPR-KKR) package [3]. The effective Hamiltonian describing the system can be represented by two interacting contributions: one that describes the magnetic interactions and the other one taking care of the structural distortion. For the magnetic part, we have chosen the anisotropic Heisenberg model with *ab initio* exchange parameters. The structural part is described by the degenerated three-state Blume-Emery-Griffiths (BEG) model allowing for a structural transformation from the cubic (austenitic) phase to the tetragonal (martensitic) phase [4]. In our model, we consider only two variants of martensite with lattice deformation along one of axes, in spite of fact, that during cooling, the austenite may choose any of the six variants. So, in our case the cubic phase is a

double-degenerated phase. Since the Ga atoms are non-magnetic atoms, we do not take into account the Mn-Ga, Ni-Ga and Ga-Ga interactions in the magnetic subsystem. But in the structural subsystem we take into account the interactions between all atoms.

The effective Hamiltonian (1) consists of three contributions, the magnetic part is described by Eq. (2), the structural part by Eq. (3), and the magnetostructural interaction is defined in Eq. (4),

$$H = H_m + H_{el} + H_{int}, \tag{1}$$

$$H_m = -\sum_{<i,j>} J_{ij}^{mag} \mathbf{S}_i \mathbf{S}_j - g\mu_B H_{ext} \sum_i S_{iz} - K_x \sum_i S_{ix}^2 - K_z \sum_i S_{iz}^2, \tag{2}$$

$$H_{el} = -J_{el} \sum_{<i,j>} \sigma_i \sigma_j - K_{el} \sum_{<i,j>} (1-\sigma_i^2)(1-\sigma_j^2) - k_B T \ln(p) \sum_i (1-\sigma_i^2) - K_1 g\mu_B H_{ext} \sum_i \delta(\sigma_i, \sigma_g) \sum_{<i,j>} \sigma_i \sigma_j, \tag{3}$$

$$H_{int} = 2U \sum_{<i,j>} \mathbf{S}_i \mathbf{S}_j (0.5-\sigma_i^2)(0.5-\sigma_j^2) - 0.5U \sum_{<i,j>} \mathbf{S}_i \mathbf{S}_j. \tag{4}$$

Here, J_{ij}^{mag} are the exchange constants in the magnetic subsystem, $\mathbf{S}$ is the spin, H_{ext} is the external magnetic field; g is the Lande factor; μ_B is the Bohr's magneton, K_x, K_z are the anisotropy constants along x and z axis, J_{el} and K_{el} are the exchange constants in elastic subsystem in the martensitic phase and austenitic one respectively, σ_i is a microdeformation variable which represents the deformation state on each site of the lattice, where $\sigma_i = 0$ denotes the undistorted (cubic) phase and $\sigma_i = -1, 1$ the distorted (martensitic) phase with two variants, k_B is the Boltzmann constant, T is the temperature of system, p is the degeneracy factor, K_1 is the dimensional magnetostuctural constant which describes the influence of the external magnetic field on the structural subsystem, σ_g is a deformation state, whose value is that of a structural variant in the external magnetic field (the positive field H_{ext} favors deformation states coinciding with the σ_g deformation state), U is the magnetostructural constant.

With respect to the elastic part of the Hamiltonian (3), the first and second terms characterize the interaction between single strains σ_i in the tetragonal phase and cubic one. The third term characterizes the degeneracy of cubic phase (we consider the degeneracy of the cubic phase equal $p = 2$). Finally, the fourth term describes the influence of a magnetic field on the subsystem [5]. The Magnetostructural contribution (4) characterizes the interactions between magnetic and structural degree of freedom.

The corresponding equilibrium Monte Carlo simulations have been carried out by means of the Metropolis algorithm [6]. For application in the systems with structural and magnetic interactions it is fully described in the reference [5].

Calculation details

Since Monte Carlo method has the stochastic nature, we have done the independent series of calculations using the same model parameters. The number of series was equal of $N = 8$. For each case the magnetic field dependences of microdeformation, spin projection and their volume fractions (x_i) at the constant temperature have been calculated. After that all calculated data have been averaged over eight series ($\bar{x}$). Also we calculated the standard deviation of calculated dependences by following way:

$$S = \sqrt{\sum_{i=1}^{n} \frac{(x_i - \bar{x})^2}{N-1}} \tag{5}$$

The Monte Carlo simulations of the temperature dependences of the magnetization and deformation were carried out for two cases. In the first case, calculations have carried out in the austenite (cubic) phase. In the second case, the calculations have carried out in the martensitic (tetragonal) phase with two martensite variants.

So, for the temperature dependences of magnetic and structural order parameters, the calculations have started with follow initial configurations. In the structural subsystem the microdeformations for all atoms we have taken of $\sigma_i = 1$, whereas in the magnetic subsystem the spin projections for the Ni and Mn atoms we have taken of $S_x = 0$, $S_y = 0$, $S_z = 1$ and $S_x = 0$, $S_y = 0$, $S_z = 0$ for the Ga atoms, respectively.

For the twin motion modeling under influence of the external magnetic field at the constant temperature, the Monte Carlo simulations have started from five lattice blocks (the size of each lattice block is C) with different configurations of spins and strains: for $z \in$ ([1, 1/5×C], [2/5×C, 3/5×C], [4/5×C, C] we have taken $\sigma_i = 1$ for all sites, and $S_z = 1$, $S_x = 0$, $S_y = 0$ for magnetic sites and $\mathbf{S} = 0$ for non-magnetic sites; for $z \in$ ([1/5×C, 2/5×C], [3/5×C, 4/5×C] we have taken $\sigma_i = -1$ for all sites, and $S_z = 0$, $S_x = 1$, $S_y = 0$ for magnetic sites and $\mathbf{S} = 0$ for non-magnetic sites.

The motion of twin boundary has created by following way. At the constant temperature and given external magnetic field in each lattice plane (101) after 1000 Monte Carlo steps the average deformation of layer is fixed. If the average deformation of layer is in the interval from -1/3 to -1 then the plane does not moved; if it is in the interval from -1/3 to 1/3 then the plane shifted along axis [101] on the distance which is equal half of diagonal of elementary lattice face in the plane (*x*, *y*); finally, if it is in the interval from 1/3 to 1 then the plane shifted along axis [101] on the distance which is equal diagonal of elementary lattice face in the plane (*x, y*). After this we have changed the external magnetic field and repeated the Monte Carlo cycle. So, using proposed trick, we can model the reorientation of martensitic variants and spins projections to the energetically favorable ones under influence of external magnetic field. Hence the twin boundary motion is appeared in the tetragonal lattice.

Calculations have been performed for three different values of anisotropy constants, which are equal (in meV) for each site type:

$$\begin{aligned}
&1.\ K_x^{Ga} = 0,\ K_z^{Ga} = 0,\ K_x^{Ni} = 0,\ K_z^{Ni} = 0,\ K_x^{Mn} = 0,\ K_z^{Mn} = 0;\\
&2.\ K_x^{Ga} = 0,\ K_z^{Ga} = 0,\ K_x^{Ni} = 0.72,\ K_z^{Ni} = 0.36,\ K_x^{Mn} = 0.18,\ K_z^{Mn} = 0.09;\\
&3.\ K_x^{Ga} = 0,\ K_z^{Ga} = 0,\ K_x^{Ni} = 0.36,\ K_z^{Ni} = 0.18,\ K_x^{Mn} = 0.09,\ K_z^{Mn} = 0.045.
\end{aligned} \tag{6}$$

The values of anisotropy constants were estimated from available experimental data for Ni-Mn-Ga alloys [7].

Simulation results

The Monte Carlo calculations have been performed on two lattices. The first lattice has the size $N = a \times a \times 4a$, where $a = 6$ is the number of unit cells of Heusler alloys. So in this case we have used the lattice with 4225 Mn, 7056 Ni, and 4225 Ga atoms. The second lattice has the size $N = a/3 \times a/3 \times 4a$. In this case we have used the lattice with 625 Mn, 784 Ni, and 625 Ga atoms. The influence of size lattice on the twin boundary motion was studied on lattices with size $N = a \times a \times 2a$ and 2112 Mn, 3456 Ni, and 2113 Ga atoms and $N = a \times a \times 5a/6$ with 845 Mn, 1296 Ni, and 845 Ga atoms.

The magnetic exchange integrals have been taken from our *ab initio* calculations using SPR KKR package: $J_{Ni\text{-}Mn} = 4.74$ meV, $J_{Mn\text{-}Mn} = 1.63$ meV for nearest neighbors sites located in *xy* plane, $J_{Mn\text{-}Mn} = -1.26$ meV for neighbors sites located in *xz* and *yz* planes [5]. In the structural subsystem the exchange interactions have been taken for all type of atoms as in our previous works: $J_{el} = 1.5$ meV, $K_{el} = 0.5$ meV, $U = 3$ meV, $K_1 = 0.5$ meV [5, 8]. The Lande factor was taken as $g = 2$.

Both the Curie temperature and the martensitic transition one of Ni_2MnGa with cubic lattice have been estimated from temperature dependences of magnetization and deformation at zero magnetic field [9]. Our theoretical results are closed to experimental data for Ni_2MnGa alloy [10].

The kinetic of twin motion and magnetic field dependences of the volume fraction of martensitic variants and volume fraction of spin projections have been investigated using the tetragonal lattice in the first case at the constant temperature $T = 120$ K and different sets of anisotropy constants. In the second case we have taken the different temperatures and lattice sizes with zero magnetic anisotropy. The calculations have been performed with the number of Monte Carlo steps equal 1000.

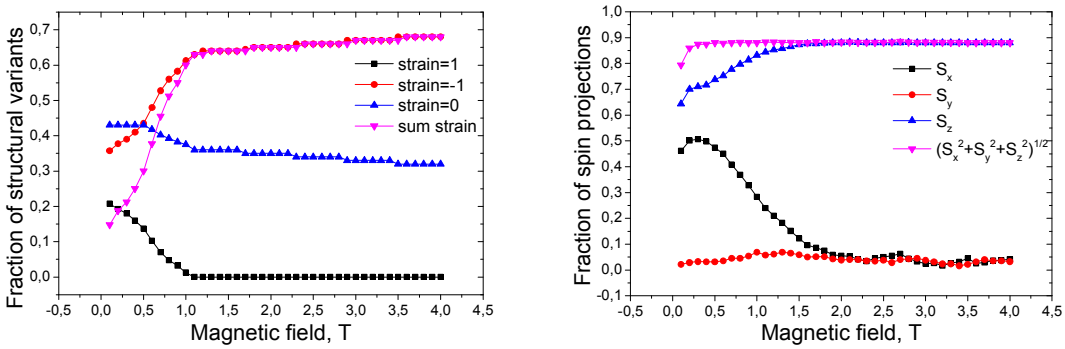

Fig. 1. Magnetic field dependences of volume fraction of martensitic variants (left) and volume fraction of spin projections (right) at the temperature T = 120 K and the first set of anisotropy constants (6).

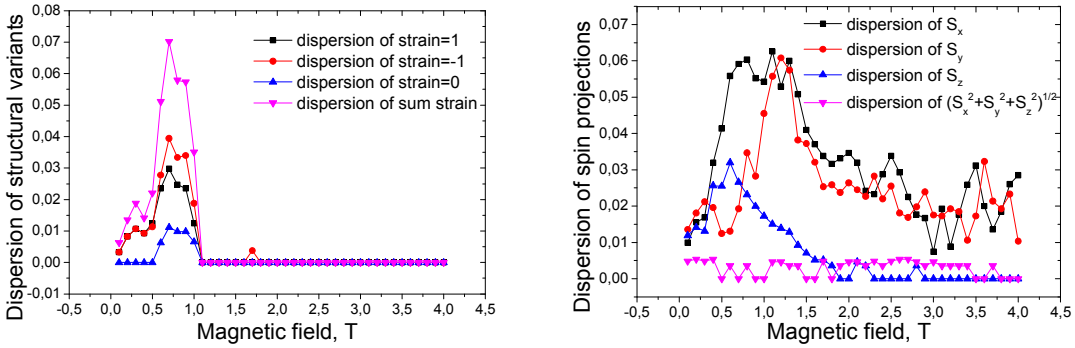

Fig. 2. Magnetic field dependences of dispersions of fraction of martensitic variants (left) and fraction of spin projections (right) at the temperature T = 120 K and the first set of anisotropy constants (6). The dispersions have been calculated using Eq. (5).

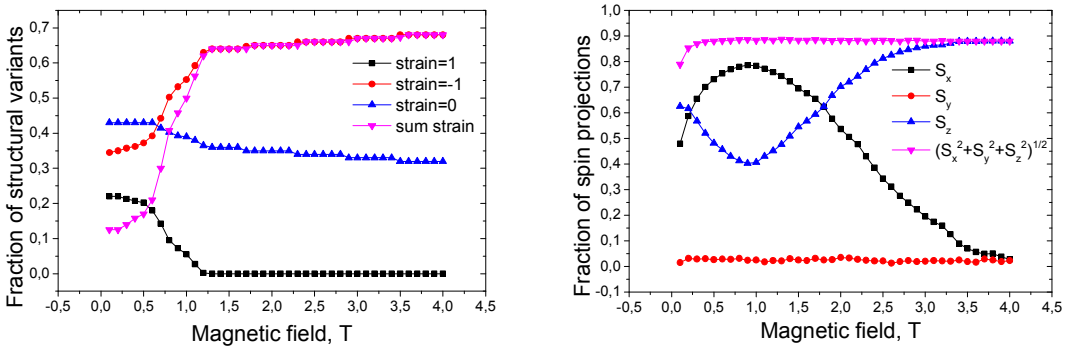

Fig. 3. Magnetic field dependences of volume fraction of martensitic variants (left) and volume fraction of spin projections (right) at the temperature T = 120 K and the second set of anisotropy constants (6).

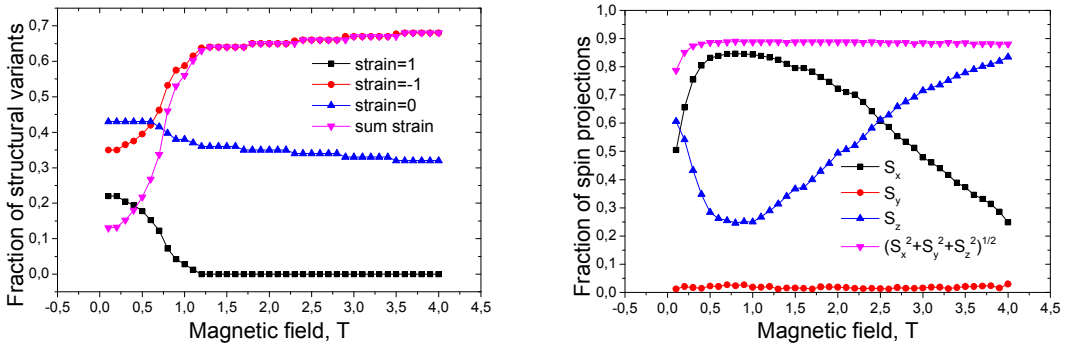

Fig. 4. Magnetic field dependences of volume fraction of martensitic variants (left) and volume fraction of spin projections (right) at the temperature T = 120 K and the third set of anisotropy constants (6).

In the Fig. 1,3,4 we present magnetic field dependences of volume fraction of martensitic variants (left) and volume fraction of spin projections (right) at the temperature T = 120 K, calculated for lattice N = 6×6×24 cubic unit cells. We can see, that increasing of anisotropy in magnetic subsystem leads to competition between x and z spin projections and to delay reorientation of the spin projection along the direction of the external magnetic field and almost no effect on transition in elastic subsystem.

In the Fig. 2 we show magnetic field dependences of dispersions of fraction of martensitic variants (left) and fraction of spin projections (right) at the temperature T = 120 K. We can see that dispersion of all types of strains in the structural subsystem after phase transition in fact equal zero (although there may be small deviations due to thermal fluctuations as on H=1.7 T), while in magnetic subsystem after phase transition dispersion of z spin projection tends to zero under influence of external magnetic field and dispersions of x and y spin projections remain nonzero due to thermal fluctuations A similar behavior of dependences of fraction of martensitic variants and spin projections is observed for all presented here dependences of fraction of martensitic variants and fraction of spin projections from external magnetic field.

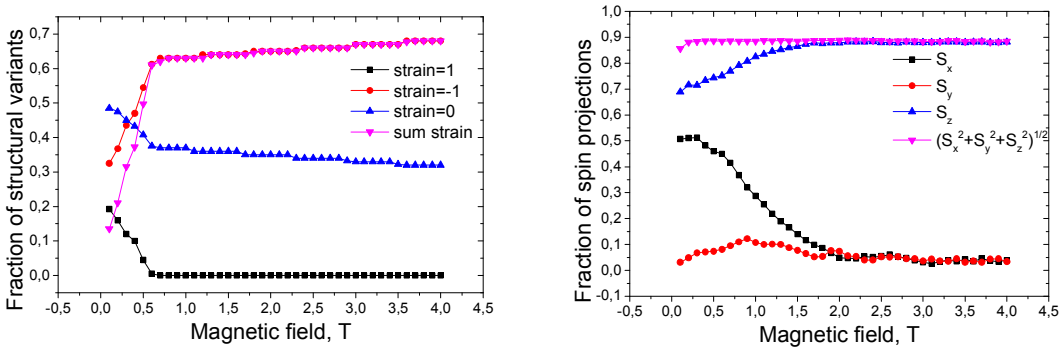

Fig. 5. Dependences of fraction of martensitic variants (left) and fraction of spin projections (right) from external magnetic field at temperature T = 120 at first set of anisotropy constants (6) for N = 6×6×12 cubic unit cells.

The fig. 5 shows, that decreasing of lattice size (from N = 6×6×24 cubic unit cells to N = 6×6×12 cubic unit cells) leads to acceleration of phase transitions in elastic and magnetic subsystems due to decreases total number of sites with energy unfavorable martensitic variants and directions of spins.

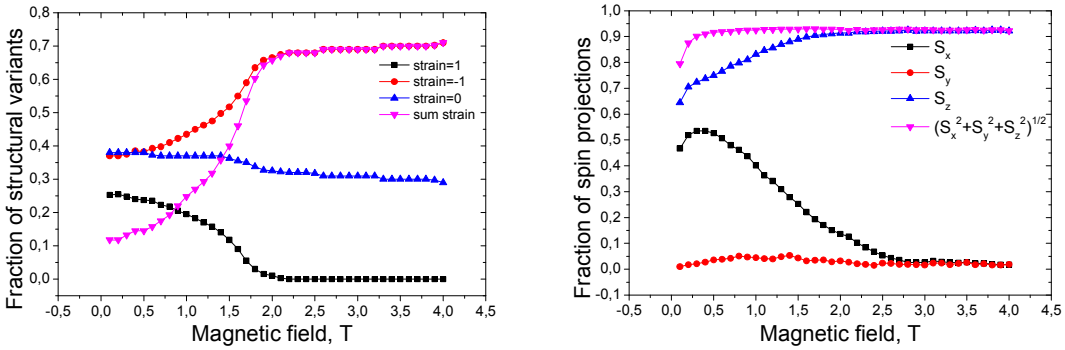

Fig. 6. Dependences of fraction of martensitic variants (left) and fraction of spin projections (right) from external magnetic field at temperature T = 80 at first set of anisotropy constants (6).

The fig. 6 shows, that reduction of temperature (from T=120 K to T=80 K) leads to slowdown of phase transitions in elastic and magnetic subsystems and speed of twins boundary moving due to decrease of thermal fluctuations in system.

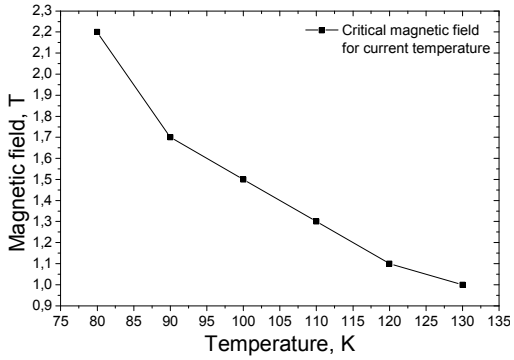

Fig. 7. Dependance of critical magnetic field in elastic subsystem for different temperatures for first set of anisotropy constants (6).

The fig. 7 shows, that value of critical magnetic field for elastic subsystem and speed of twins boundary moving decrease with temperature growning due to increase of thermal fluctuations in system. Critical values of magnetic field for different temperatures obtained from dependence of dispersion of fraction of martensitic variants from external magnetic field as values, at which dispersion of summary microdeformation of lattice turns to zero.

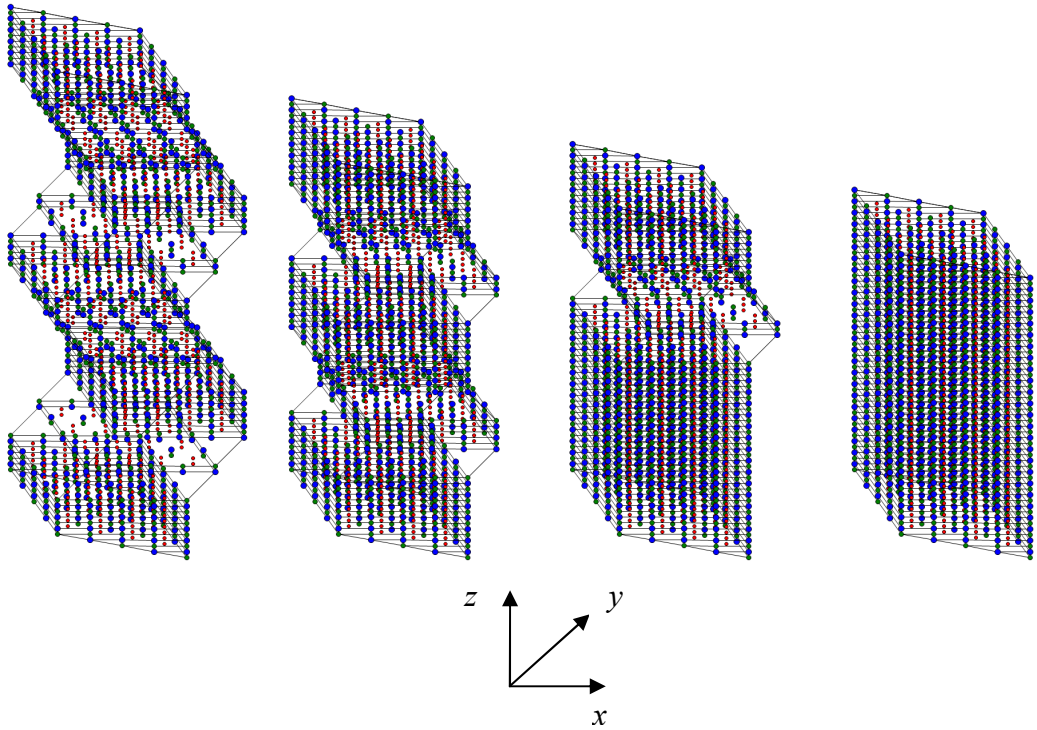

Fig. 8. Image of lattice at external magnetic field (from left to right) H=0 T (starting configuration), H=1 T, H=1.1 T, H=1.2 T.

The fig. 8 show the kinetic of twin motion under influence of external magnetic field at constant temperature T = 120 K. Here small circles corresponds the atoms of Ni, medium circles denotes the atoms of Ga, big circles are the atoms of Mn. Note that temperature have significant effect on the system, what leads to fluctuation and changing values of spins projections and microdeformations and to changing normalized microdeformation of layers and their spacing.

Summary

In this work we have proposed the microscopic model based on the real lattice of Ni-Mn-X (X=Ga, In, Sn, Sb) alloys and classical Heisenberg and Blume-Emery-Griffiths models with magnetoelastic interaction for description of the twin boundary motion in Ni_2MnGa alloy under

influence of an external magnetic field. On the base of this model the temperature dependences of magnetization and microdeformations at zero magnetic field have been investigated. The kinetic of twin boundary motion under influence of external magnetic field at constant temperatures has been studied with the help of proposed model. Our simulations have shown that an applying of the external magnetic field at constant low temperatures leads to moving of twin boundary. The motion of twin boundary undergoes during transformation an unfavorable martensitic variant in another variant, which has the same direction as an external magnetic field. We have found that the velocity of twin motion depends on the external magnetic field and temperature. It should be noted that the theoretical investigation of twin boundary motion agree with experimental results [11, 12].

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