

## Time-dependent effects caused by the magnetic field in the Ni–Mn–Ga magnetic shape memory martensites

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Present paper is a review of published and new original results concerned on study the phenomenon of the time-dependent evolution of the magnetic-field-induced strain under constant magnetic field at fixed temperature founded recently. The time dependent evolution of martensitic structure in the constant magnetic field, stability of martensite under the magnetic field, time-dependence of the magnetic field induced strain and magnetization in the Ni–Mn–Ga magnetic shape memory martensites are studied in single and polycrystals. An analogy in the time evolution of the magnetically and mechanically induced deformations in the stationary conditions is shown. Effects of temperature, the magnetic field magnitude and its direction are analyzed. Physical processes responsible for the time dependent evolution of martensitic structure and correspondently the magnetic field induced strain under stationary conditions are analyzed. Mechanisms of observed time-dependent evolution of martensite structure and strain in a constant magnetic field are proposed. It is shown basing on the theory of random processes that thermal fluctuations of microstresses caused by the constant field is responsible for the time-dependent magneto-mechanical behavior of the Ni–Mn–Ga magnetic shape memory martensites. Activation of the new twinning systems and their evolution determines the long-time evolution of martensite in the stationary conditions in the case of application of the magnetic field to the hard or some arbitrary direction of magnetization.

Статья является обзором опубликованных и новых результатов исследований явления зависящей от времени магнитоиндуцированной деформации под действием магнитного поля фиксированной величины при постоянной температуре. Исследована временная эволюция мартенситной структуры в фиксированном магнитном поле, временная зависимость магнитоиндуцированных деформаций и намагнитченности в Ni–Mn–Ga мартенситах с магнитной памятью формы. Показана аналогия во временной зависимости деформаций под действием фиксированного магнитного поля и механических напряжений постоянной величины. Исследовано влияние на деформацию температуры, величины и направления магнитного поля. Проанализированы физические процессы, ответственные за временную эволюцию мартенситной структуры и, соответственно, деформации в стационарных условиях в магнитном поле фиксированной величины и при постоянной температуре. Предложены механизмы временной эволюции мартенситной структуры и деформаций под действием постоянного магнитного поля. Базируясь на теории случайных процессов, показано, что термические флуктуации микронапряжений, создаваемых магнитным полем, ответственны за временную эволюцию магнито-механического поведения Ni–Mn–Ga мартенситов с магнитной памятью формы. В случае соответствия направления магнитного поля "жесткой" оси намагнитченности в мартенсите с пятислойной модулированной структурой, активация новых систем двойникования и их развитие ответственно за долговременную эволюцию мартенситной структуры в стационарных условиях.

As it is known, a giant magnetically-induced deformation of Ni–Mn–Ga martensites was discovered not so long ago ([1–6] and

the others) and denominated as magnetostrain effect (MSE) and/or magnetic shape memory effect (MSME). The MSE effect is

caused by the reorientation of the twinned martensitic variants under the action of external magnetic field applied to the specimen. In the case of quasi-stoichiometric Ni-Mn-Ga alloys with  $c/a < 1$  ( $c$ ,  $a$  are the lattice parameters of the tetragonal *ft* martensite [2–6]) the strain arises due to the increase of the volume fraction of martensite variant with  $c$ -axis parallel to the field and therefore, is characterized by the contraction of the single crystal in the field direction and elongation in the  $a$ -direction.

A slow variation of the magnetically-induced strain  $\varepsilon(H)$  and effect of time exposure was reported firstly in the [7] for the Ni-Mn-Ga alloy exhibiting comparatively small magnetostrain effect with  $\varepsilon(H_S) \sim 0.1\text{--}0.2\%$  ( $H_S$  is a saturation field). A long-time evolution of the martensitic structure and correspondent magnetically-induced strain was observed and studied experimentally in the martensitic single crystal displaying the magnetostrain of about of few per cents [8–11]. First theoretical investigations of time-dependent phenomenon in constant magnetic field were introduced very recently [12, 13]. It was shown, that the magnetically-induced deformation varies noticeably within the wide interval of time (from a fraction of a second to the few hours or days, depending on the experimental conditions and alloy specimen). It is quite obvious that the studies of the time evolution of the magnetically induced deformation is of a special importance in view of the expected applications of the Ni-Mn-Ga alloys as the new elements of magneto-mechanical transformers, actuators and controllers.

Present paper is dedicated to the systematic review and physical interpretation of the time-dependent effects caused by the magnetic field under the stationary conditions in the magnetic shape memory martensites of Ni-Mn-Ga. A special attention is paid to the clarification of mechanisms of time-dependent evolution of martensitic structure and correspondent strain and its interpretation as the internally stressed state.

Single crystal specimens displaying magnetic field induced strain more than 5 % at room temperature in elevated magnetic field were studied. Chemical compositions of alloys are showed in the Table.

Various methods of studies were used: tensometry in the magnetic field, neutron diffraction, X-ray diffraction (theta-two theta method and texture analysis) and an optical microscopy. All experiments con-

cerning study of the time-dependent behaviour in the external magnetic field were performed "in-situ". Details of the specimen's preparation and experimental procedures are explained in [7–11]. The single crystalline specimens with surfaces parallel to the {100} crystal planes were preliminary magnetized in the field 0.5 T or 0.9 T in the direction coincides with the [001] (direction of easy magnetization) in one of the twinned variants. Then magnetic field was applied in the perpendicular direction during the experiments for study effect of the magnetic field on strain or the structure evolution. Strain was measured perpendicular to the applied field direction. Strain in the magnetic field produced by electromagnets was measured by using an uniaxial magnetic dilatometer [14]. An optical microscopy study of the martensite structure evolution in the steady magnetic field is carried out using the Nd-Fe-B permanent magnets. Experimental procedure of the neutron diffraction study is explained in [15, 16].

*Structure of magnetic shape memory martensites.* It is well known that crystal structure of martensite in Ni-Mn-Ga alloys depends strongly on compositions. The main types of the crystal structure of martensite existing in bulk specimens of Ni-Mn-Ga Heusler alloys are *bct* non-modulated, 5M, 7M and they're mixture. Our experience in study of the magnetic shape memory in Ni-Mn-Ga and some other published results convinced us that only martensites having 5M or 7M crystal structure type display giant magnetic field induced strain. Fig. 1 represents some examples of the crystal structure study in the magnetic shape memory martensites studied in the present work using diffraction methods. The reciprocal space (insert in the Fig. 1) for the martensite A (Tabl) represents result of neutron diffraction study [15, 16]. Additional spots located between main spots characterize the

Table. Chemical compositions and transformation temperatures in the studied single crystals

Name	Content (at. %)			$M_s$ , K	$A_s$ , K	$T_C$ , K
	Ni	Mn	Ga			
A	48.6	30.16	21.3	300	304	368
B	49.7	28.7	21.6	306	310	372
C	48.2	30.8	21.0	307	315	370
D	49.6	28.4	22.0	305	314	373

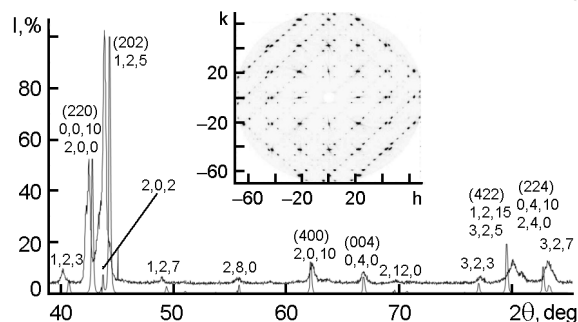


Fig. 1. Crystal structure of magnetic shape memory martensites as result of study with diffraction methods: X-ray diffraction pattern of martensite of powder specimen prepared from the single crystal C. Indication of peaks is given for 5M modulated structure and for its approximation by *fcc* unit cell (bold). The reciprocal space ( $l = 0$ ) for 5M martensite in the single crystalline  $\text{Ni}_{48.57}\text{Mn}_{30.14}\text{Ga}_{21.4}$  according to the neutron diffraction data at  $T = 297$  K; indexes  $hkl$  are given in terms of *fcc* structure is shown in the insert.

close-packed planes with periodicity of 5 lattice parameters and indicate the 5-layered modulated structure. The unit cell of martensite lattice is determined as orthorhombic 5M with lattice parameters:  $a' = 0.421$  nm,  $b' = 0.562$  nm,  $c' = 2.105$  nm at 296 K. Fig. 1 shows powder diffraction pattern for martensite of alloy C. In this case martensite also has 5M modulated structure. Structure of that martensite was determined in [17] as 5-layered monoclinic with cell parameters:  $a = 0.4225$  nm,  $b = 0.5577$  nm,  $c = 2.1030$  nm and  $\beta = 90.3^\circ$  at  $T = 296$  K.

Because MFIS is limited by the tetragonality [2–6] or by the lattice distortion  $1 - c/a$  that is confirmed by many experimental results published by different authors, it is convenient to simplify description of the real 5M crystal lattice in the magnetic shape memory martensite as tetragonal cell by ignoring of the 5-layer structure. The martensite lattice can be approximated by *fcc* cell following by [3–6, 15 and others authors] relating to the parent *fcc* austenite phase (Fig. 1). In that case martensite lattice parameters for alloy A are:  $a = b = 0.595$  nm and  $c = 0.561$  nm at  $T = 296$  K in the terms of *fcc* cell. The shifting system is  $(110) [1-10]_{fcc}$  for this *fcc* lattice. Crystallographic relation between *fcc*  $\mathbf{a}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$  axes and  $\mathbf{a}'$ ,  $\mathbf{b}'$ ,  $\mathbf{c}'$  axis in 5-layered modulated structure are given in [17]. Indexes for 5-layered orthorhombic

structure are transformed in case of *fcc* cell as it is shown in the Fig. 1:  $(105)_{5\text{-layerd}} \rightarrow (200)_{fcc}$ ,  $(020)_{5\text{-layerd}} \rightarrow (002)_{fcc}$ . Indexes (1 2 7) and (2 0 8) belonging to the 5-layered structure are ignored for the approximation by *fcc* lattice. We also will use description of a martensite in terms of *fcc* tetragonal lattice in the beginning of the present paper for simplicity as is the convention, however will show below, that this kind of approximation does not explain some important physical processes which are responsible for the magnetic field induced strain and its time dependence.

*Evolution of the magnetic field induced strain in steady magnetic field at constant temperature.* Fig. 2a shows an examples of the magnetic field induced strain in the elevated magnetic field in the martensites of single crystals A and B (Tabl.). MFIS in the martensite is caused essentially by a re-alignment of twins [2–6, 8, 10], so there is an excessive proportion of the *c*-axis contribution to the dimensions of the specimen along the direction of the applied field. Ratio  $c/a < 1$  in the *fcc* approximation of crystal structure, hence the specimen is contracted in the direction of the applied magnetic field, because [001] martensitic variant with its *c* short axis is oriented to the applied field. In the same time the single crystal specimen rises in size in the *a* direction normal to the field, which consists with the direction of strain measurement (Fig. 2a). After this experiment the specimen was re-magnetized in the perpendicular direction and thereafter the magnetic field was again applied quickly up to 0.6 T (Fig. 2b) in the same direction as in the case presented in the Fig. 2a. Measurement of strain was continued after the magnetic field stabilization. As follows from the Fig. 2b the stain grown at 1.7 % during 52 min in steady magnetic field 0.6 T without saturation. The same increment of strain is achieved due to rise in the magnetic field up to 1 T in the case of the quick increase magnetic field (Fig. 2a). In some cases (Fig. 2d) strain achieves large magnitudes 3.6 % during the short exposure (10 min) in the small field 0.48 T (Fig. 2d), displaying the overall strain 5 % that is equal to the strain in the elevated up to 1 T magnetic field (Fig. 2d).

Value of strain caused by the constant magnetic field can be quite small, or much bigger during the same time exposure in the magnetic field Fig. 2b, 2d and [7–11]. Time-dependent phenomenon is observed as for

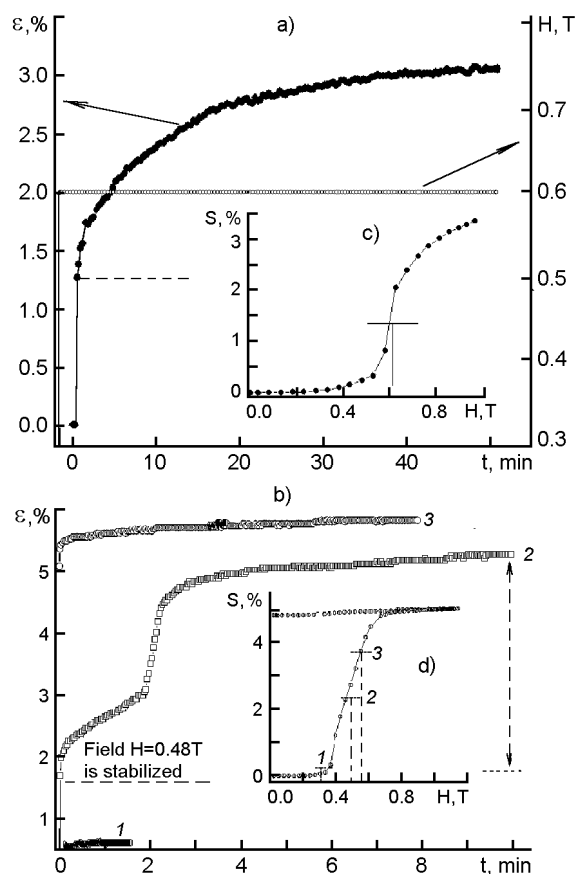


Fig. 2. Strain induced by the magnetic field in the martensitic single crystals at constant temperature: (a) strain in the evaluated magnetic field, single crystal A; (b) time dependent strain in the constant magnetic field 0.6 T, single crystal A; (c) magnetically induced strain in the quickly (1, 2 min) applied elevated field for single crystal B; (d) complete strain value in the quickly applied magnetic field and its' rise with time after the field stabilization, crystal B.

poly- as well for single crystals Ni-Mn-Ga displaying the magnetic shape memory [7–10]. It was shown with X-ray diffraction and optical microscopy study [7–10] that time-dependent evolution of the twinned martensite and twins' redistribution under steady magnetic field responses on the time-dependent strain. Defects (dislocations, vacancies, other boundaries and their intersections cause pinning effect and bloke twin boundaries. Density of the pinning centers is various in different specimens and depends on their history, therefore as the magnitude of time dependent strain, as well as its kinetics is different. However, there are common tendencies and regularity in the time-dependent behavior in the constant

magnetic field, namely: the magnetic field value, effect of temperature, crystallographic direction of the applied magnetic field. These common tendencies for the time-dependent behavior in the magnetic shape memory martensites are analyzed below.

*Resemblance in an effect of the magnetic field and mechanical stress on the time-dependent behaviour of ferromagnetic martensites.* There is an analogy in the strain of the Ni-Mn-Ga crystal in the elevated magnetic field with that under applied mechanical stress applied to the same crystallographic direction for the same single crystal (Fig. 3a). Martensite gradually deforms under constant magnetic field and set temperature in a manner analogous to creep under the constant applied compressive stress (Fig. 3b). therefore. Basing on founded analogy with mechanical creep, the time-dependent strain in constant magnetic field at fixed temperature was denoted as "magnetic creep" [11, 9].

Magnetic field-strain in the elevated magnetic field and stress-strain curves, were analysed in a few articles [for example 3–6, 18–22 and references therein]. Authors [19] have shown that there is an analogy between strain caused by mechanical and by magnetic driving forces for twin boundary motion in martensite Ni-Mn-Ga. The resemblance of the field-induced and stress-induced strain (Fig. 3a) allows to refer to the equivalence principle, which was stated earlier [20, 21] for the magnetic and mechanical stressing of the shape-memory alloys (see also [22] and references therein). In the present article we accent on some peculiarities of strain behaviour, which are important for analysis the time-dependent phenomenon that is a subject of present investigation because it is assistes to underset of an a physical nature the magnetic creep.

There are some critical magnitudes as for the magnetic field caused quick response of the field/stress relaxation. Strain rises quickly and more notably under field/stress value close to these critical values (Fig. 3a). Thereafter strain rises slightly due to the increase the magnetic field/stress value. The lattice distortion limited theoretical maximum for the single crystal A is  $(1-c/a) \cdot 100 = 5.7\%$  at 293 K [16]. Big values of strain achieved under small field/stress magnitudes and almost maximal values of field/stress induced strain at used temperature, indicates that

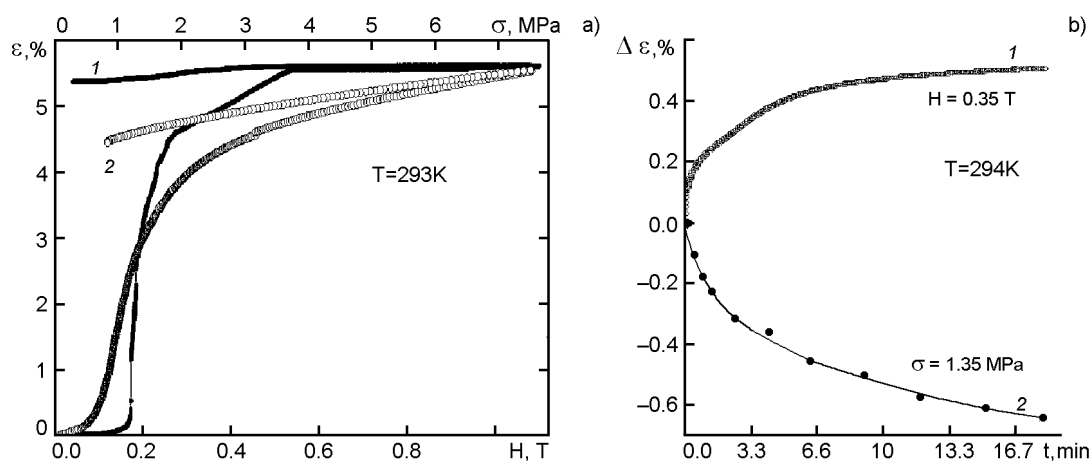


Fig. 3. Analogy in the magnetic field induced strain and stress-strain behavior in the single crystal A: (a) strain induced by the elevated magnetic field (1) and mechanical stress (2); (b) time-dependent strains induced by the stationary magnetic field 0.5 T (1) and mechanic stress 0.55 MPa (2). It is shown a module of the strain magnitude caused by the compressive mechanical stress.

used single crystal has quite small density of defects blocked twin boundary motion. Rise in the field/stress value causes de-pinning effect of twin boundaries and strain rises until saturation (Fig. 3a). Fig. 3b demonstrates resemblance in the time evolution of the magnetically and mechanically induced deformations in the stationary magnetic field and under the constant mechanical loading at fixed temperature for the same specimen. Firstly an analogy in the time-dependent behavior under fixed magnetic field at room temperature with that under constant mechanical stress was found in [11]. Magnitudes of the field and strain (Fig. 3b) were both chosen to be correspondent to the slow rise of strain in the elevated field and stress (Fig. 3a). Strain of about 1–1.75 % arises momentary on the quick field/stress application and then the deformation smoothly increases with the values exceeding 0.5–0.6 %. We can see that not only rise in stress or the magnetic field causes de-pinning effect, but the time exposure in the constant field/stress also.

*The theoretical interpretation of the time-dependent phenomenon.* The first attempt of a theoretical interpretation of this effect was made in [12]. As a result, the following physical picture of time-dependent MSE in the Ni–Mn–Ga magnetic shape memory martensites was substantiated:

(i) a magnetic field application results in an internal magneto-mechanical stressing of martensitic alloy specimen with stresses being different for the differently oriented martensite variants, and therefore, a microstressed martensitic state arises;

(ii) quick partial relaxation of the microstresses takes place due to the displacements of mobile coherent interfaces, thus, the quick magneto-mechanical response arises;

(iii) the quick relaxation of the field-induced microstresses is not complete because of imperfections of the crystal lattice and the incoherent character of some interfaces;

(iv) thermal fluctuations of the residual microstresses result in its slow relaxation and the relevant slow increase of the field-induced deformations up to saturation.

The computing of processes based on the theory proposed shows (Fig. 4) that the strain evaluated with time in the constant magnetic field does not reach the saturation even after one year and more, how it was computed in [12] and is shown in the Fig. 8.

The next step in the theoretical explanation of time-dependent phenomenon we talk about, was made in [13]. The conception of slow magneto-mechanical response [13] is specified as the deformation, which follows the magnetic field application after the period of time exceeding by the order of magnitude a characteristic period of transversal sound. The deformation arising before this period of time is conventionally attributed to the moment of the field application and is referred to as the quick magnetoelastic response. A statistical model describing the time-dependent phenomenon of strain rise in the constant magnetic field has been deduced from the mathematical theory of random processes [24]. We concluded [13] that thermal fluctuations of microstresses strongly affect the magneto-mechanical of

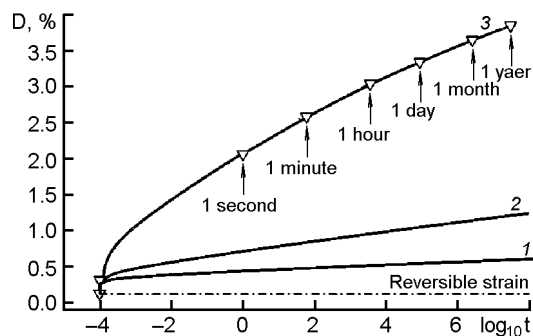


Fig. 4. Magnetic-field-induced strain versus the common logarithm of time [13]. Curves are computed for magnetic field  $H = 0.35$  T.

1 -  $\varepsilon_p = 0.15$  MPa; 2 -  $\varepsilon_p = 0.3$  MPa; 3 -  $\varepsilon_p = 0.6$  MPa.

behavior of the magnetic shape memory martensites in Ni-Mn-Ga alloys. The relaxation of fluctuating microstresses and the tendency to the minimization of magneto-static energy determines an origin of the discussed time-dependent phenomenon. Elaborated theory [13] suggests a jump-like magneto-mechanical response of martensite due to the randomly occurred micro stress fluctuations causing de-pinning. This theory is in the good agreement with the jump-like behaviour in the time-dependent strain in the constant magnetic field (Fig. 5a) observed experimentally. Jump-like deformation occurs quickly (at  $<0.1$  sec) at exposure in the constant magnetic field at constant temperature. The effects of this kind point to the avalanche displacement of the

different elements of martensite microstructure caused by the concentration of local microstresses in some domain of the specimen. The concentration of microstresses can be conditioned, in its turn, by the creep of dislocations and point defects of the crystal lattice.

One of the important effects of time-dependent behavior of the magnetic shape memory martensites is rise of magnetization with exposure at fixed temperature in the constant magnetic field (Fig. 5b). Taking into account correspondence of  $90^\circ$  magnetic domains with twins [10] in studied martensites and analogy in the time-dependent behavior of strain and magnetization including jump-like changes (Fig. 5a, b), we can conclude, that origin of rise in magnetization with time is determined also by the time-dependent movement of twins oriented by easy axis of magnetization to the applied field. However clarification of details in the time-dependent changes in the domain magnetic structure in the constant magnetic field requires some additional study.

*Effect of temperature.* The effect of temperature on MFIS under the elevated magnetic field is reported in [17,25,]. It was shown that rise in temperature causes decrease of the critical magnetic field needed for twin boundary motion. Therefore we can expect an influence of temperature on time-dependent behaviour of twins in the stationary field. Really, the magneto-mechanical response to the fixed magnetic field slows down with the temperature decrease (Fig. 6). Stress relaxation occurs more

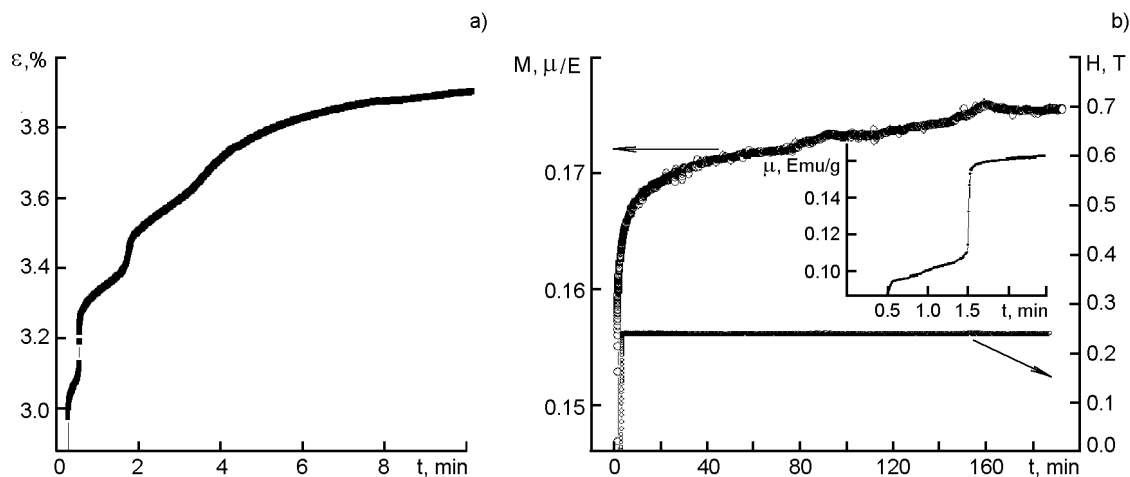


Fig. 5. Jump-like magnetically induced strain and magnetization in the constant magnetic field at fixed temperature in martensite of alloy C: (a) time-dependent strain; (b) magnetization. Insert shows the jump-like behavior of magnetization.

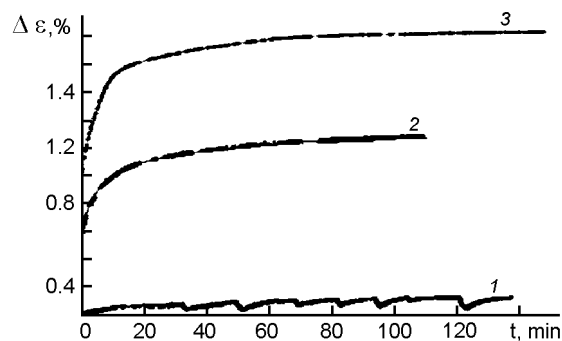


Fig. 6. Effect of temperature on the time-dependent strain in martensite of the alloy A in the constant magnetic field. 1 -  $T=273$  K; 2 -  $T=293$  K; 3 - 303 K.

quickly at higher temperature. Temperature dependence of redistribution of twins in the magnetic field, as well as effect of temperature on the critical magnetic field value as well as, is studied in-situ with neutron diffraction in [15]. It was shown that a value of the martensitic variant oriented its easy axis normal to the applied field decreases more intensively at higher temperature. Effect of elevating temperature on the acceleration of the time-dependent processes responding on strain can be caused by following reasons: well known decreases of the activation energy for twins' dislocations and higher mobility of twins at higher temperatures; intensification of thermal fluctuations [13] arising from the temperature increase causes de-pinning of twins more intensively and enabled jumps of the de-pinned boundaries.

*Effect of the magnetic field magnitude.* Magnitude of the constant magnetic field also notably affects on the time-dependent evolution in strain at fixed temperature [8,9,11] (Fig. 2d). Strain evaluated with time markedly if the magnetic field magnitude is close to the critical magnetic field value (Fig. 2c, d, Fig. 7). If the field value are close to saturation or smaller than a critical field value, strain rises with time slowly and value of strain is less at the same exposure (Fig. 2d, Fig. 7a). Twin boundaries continue slow translation after start of movement under the constant magnetic field that is higher than critical value, required for start of twins (0.35 T in the case Fig.). Effect of the mechanical stress direction on the time-dependent strain was studied in [11]. If exposure occurs at the field or mechanical stress value higher than critical magnetic field/stress, however much smaller than saturated field/stress (Fig. 2c, d, Fig. 7a, b),

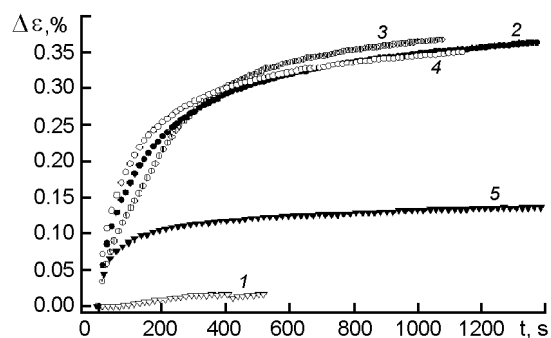


Fig. 7. Effect of magnitude of the constant magnetic field on strain in the martensite of single crystals A at 294 K. 1 -  $H=0.19$  T; 2 -  $H=0.349$  T; 3 -  $H=0.504$  T; 4 -  $H=0.643$  T; 5 -  $H=0.811$  T.

there are following factors effecting movement of twins: stress caused by the magnetic field is higher than critical stress of twinning dislocation, stress is enough to provide de-pinning effect, local stress relaxation causes the non-equilibrium magnetically deformed state and stress fluctuation responds on time-dependent strain. If the magnetic field magnitude smaller than critical magnetic field, time-dependent strain occurs only due to relaxation of the thermal fluctuation of local stress. Strain value is very small in this case. If a constant field is much more than the critical field/stress value, stress fluctuations produce small increase in strain with time, because cardinal de-pinning of twins occurred early, during the magnetic field evaluation. However, reduction of the intensity of stress fluctuations results in decrease of magnetostrain with time in all cases (Fig. 2d, Fig. 7).

*Effect of the magnetic field direction.* How it was shown with X-ray diffraction of Ni-Mn-Ga magnetic shape memory single crystals, evolution of martensite under the constant magnetic field can continue from minutes up to times hours depending from the magnetic field direction [7-10]. Redistribution of the martensitic variants occurs rapidly if magnetic field is applied to the direction of easy magnetisation in one of the twin variants and, in contrast, it takes several hours or days if the constant field is applied in the "possible" or some arbitrary direction. Ratio of martensitic variant preferable oriented to the field rises insignificantly with time if field is applied to the "easy" axis [7-10]. These results are in an agreement with study of time-dependence of strain. Results of measure time-dependent strain under field applied to the "possible"

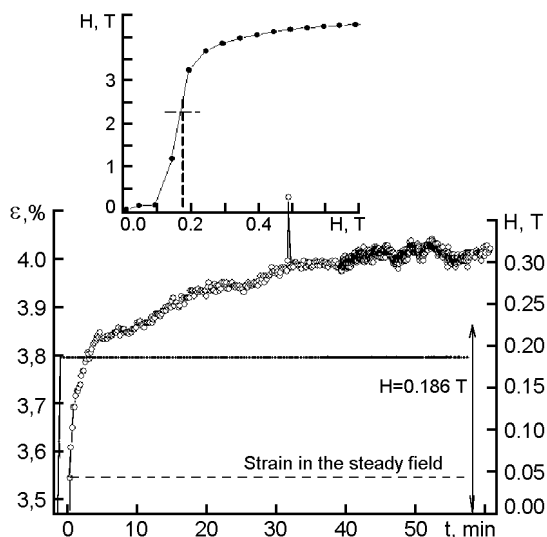


Fig. 8. Strain in martensite of single crystal A at 296 K in the magnetic field applied to the easy direction. Insert: strain in the elevated magnetic field.

and "easy" axis of magnetization for the same single crystal present Fig. 2a, b and Fig. 8, correspondently. Effect of the magnetic field direction on strain in elevated field is analyzed in the literature not enough. Some aspects of the applied field direction on strain were analysed in [26]. Critical field as well as field for the strain saturation are extremely small in the case presented in the Fig. 8a to compare with these in the Fig. 2a. These differences attracting a typical behaviour of magnetic shape memory martensites are out of the subject of present paper and will be discussed in our special article. Taking into account these differences, we choose magnitude of the constant magnetic field for study of the time-dependent strain (Fig. 8b and Fig. 2b) to be equal to the same conditions—almost middle magnitudes situated between the critical magnetic field value and field required for the strain saturation: 0.186 T (Fig. 8a) and 0.6 T (Fig. 2a). The strain evolution with time in the constant field is different for these directions. Rise in strain is 3.8 times larger if field is applied to the "possible" direction to compare with that applied to the "easy" [001] direction during the same exposure 50 min: 1.7 % and 0.45 %, correspondently (Fig. 8b and Fig. 2b). In both cases analyzed above (Fig. 2, Fig. 8) evolution of martensite is caused by movement of twins belonging to the preferable oriented variant due to de-pinning of blocked boundaries occurring with time. Re-

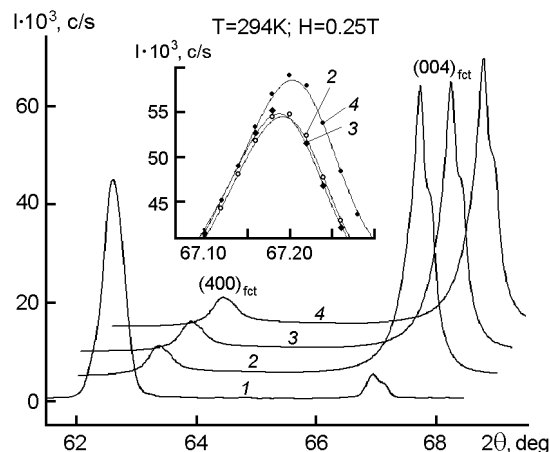


Fig. 9. X-ray diffraction pattern of single crystal D studied in-situ with exposure in the magnetic field applied to the [001] direction. Inserted fragment shows change in the (004) peak intensity during 28 hours in the magnetic field.  $\text{CuK}_\alpha$  irradiation.

distribution of twin variants with time in the constant field occurs in the same way as in the elevated magnetic field, but slowly due to de-pinning of blocked boundaries and fluctuating stresses. Value of martensitic variant expanding with exposure in the field rises insignificantly if the magnetic field is applied to the [001] easy direction (Fig. 9).

However, martensite demonstrates very peculiar behaviour in the time-dependent evolution in a case of the constant field application to the "hard" axis or to some arbitrary directions. Fig. 10 shows result of X-ray diffraction study of an effect of exposure in the constant magnetic field applied parallel to the direction  $[010]_{fct}$  on martensitic structure. Crystal had almost single variant state (010) in the studied surface before application of the magnetic field (Fig. 10a). Thereafter the specimen was clamped by its surface parallel to (010) on a permanent magnet and studied in-situ (Fig. 10b) with time. It is seen (Fig. 10a and b), that intensity of the  $(040)_{fct}$  peak decreases drastically as well as in the case shown in the Fig. 10 after field application. However in this case there is no re-orientation between  $(040)_{fct}$  and  $(004)_{fct}$  variants. Instead of that new variants are appeared. These variants can not be explained in the frame of approximation of real structure in martensite by the tetragonal  $fct$  unit cell (Fig. 1), which we used above following by others authors. These new peaks belong to the 5M modulated lattice and correspond to the (2, 8, 0) and (2, 12, 0) (Fig. 1). These



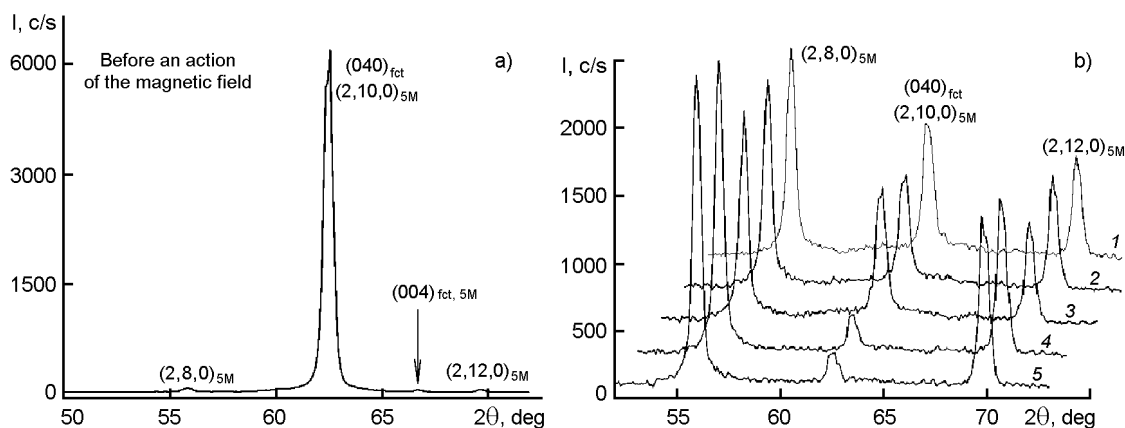


Fig. 10. Experimental  $\theta$ - $2\theta$  diffraction patterns for single crystal D at room temperature: (a) before an action of the magnetic field; (b) evolution with time of martensite in the magnetic field. Magnetic field applied normal to the studied surface.  $\text{CuK}\alpha$  irradiation is used.

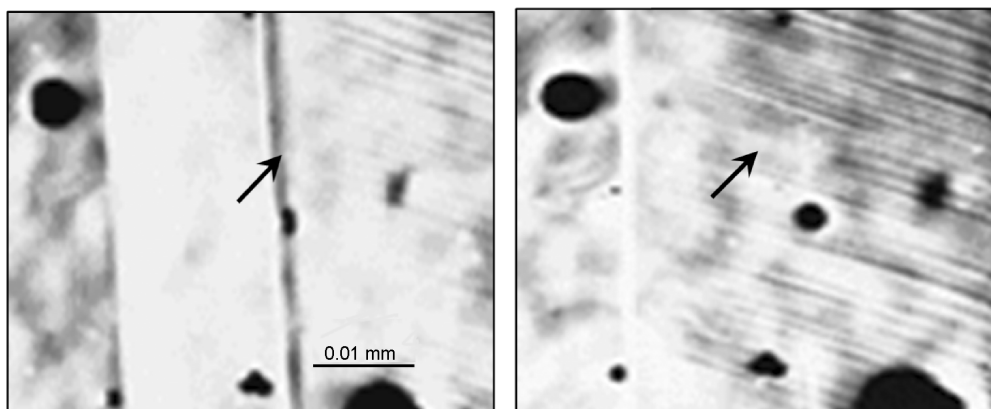


Fig. 11. Martensite structure evolution of the crystal D with exposure in the constant magnetic field 0.3 T at room temperature studied in-situ: (a) 0.37 hours in the magnetic field; (b) 17.37 hours under magnetic field. Magnetic field is directed normal to the studied surface.

orientations are twinned to the each other. Crystallography of twinning in the 5M martensite of Ni-Mn-Ga was studied firstly in [17]. Intensity of both of them rises with exposure in the magnetic field, whereas intensity of the initial  $(2, 10, 0)$  5M peak, which corresponds to the  $(040)$  for the *ftc* approximation, continues to decrease with time (Fig. 10b). Presented results (Fig. 10) show necessity to use the correct description of magnetic shape memory martensites in terms of real 5-layered modulated structure not only for the time-dependent processes, but for magneto-plasticity in Ni-Mn-Ga in the elevated field. Ignoring of the real 5M structure does not let to understand nature of processes occurring in ferromagnetic martensites in the magnetic field.

Fig. 11 shows the structure evolution with time in the constant magnetic for the case discussed just now. Experiment is performed in-situ: the specimen D lies on the

permanent magnet during whole time of the experiment. Direction of the magnetic field is normal to the studied surface Fig. 11 a demonstrates a new twins' system consisted thin parallel twins appearing in the right area of picture as result of the magnetic field application. This poly-twin develops with time as the single whole system occupying the left area (Fig. 11b). Moreover, the new poly-twin system devours the old twin boundary (arrow in the Fig. 11b) and that boundary disappears with long time exposure in magnetic field.

Results discussed above (Fig. 10 and Fig. 11) testify to existence an another mechanism of structure evolution in the magnetic field that is different from well known redistribution of two twin martensitic variants proposed in [2-6] and confirmed early for example in [8, 10, 15, 27 and others]. New mechanism concerns the structure relaxation in the magnetic field applied to the

some arbitrary direction of the magnetic field. New twinning system is activated due to stress caused by the magnetic field, forming poly-twin system containing a new two twinned orientation. We have deal in this case with mechanical twinning in the magnetic field concerning on as an origin of new twins, as well as with they're propagation. Expansion of poly-twin system occurs as in elevated magnetic field, as well as in the constant magnetic field with long-time exposure. Crystallographic aspects of the structure change and probably transformation in the crystal lattice will be discussed in our special paper.

*Acknowledgements.* The authors would like to thank Mrs. Yanling Ge (Helsinki University of Technology, Finland), professor Victor L'vov (T.Shevchenko National University, Kyiv) for participation in studies. Author greatly appreciates the financial support of European Office of Aerospace Research & Development (EOARD) in the frame of the STCU P-137 project.

### References

1. K.Ullakko, J.K.Huang, C.Kantner et al., *J.Appl. Phys.*, **69**, 1966 (1996).
2. K.Ullakko, *J. Mater. Engin. Perform.*, **405**, 5 (1996).
3. R.C.Handley, *J. Appl. Phys.*, **83**, 3263 (1998).
4. R.D.James, M.Wutting, *Phil. Mag. A*, **77**, 1273 (1998).
5. R.C.O'Handley, *J. Appl. Phys.*, **83**, 3263 (1998).
6. R.C.O'Handley, S.J.Murray, M.Marioni et al., *J. Appl. Phys.*, **87**, 4712 (2000).
7. N.I.Glavatskaya, K.Ullakko, *J. Magn. Magn. Mater.*, **218**, 256 (2000).
8. N.Glavatska, V.Gavriljuk, I.Glavatskiy et al., *J. Phys. IV (France)*, **8**, 281 (2001).
9. N.Glavatska, I.Glavatskiy, Y.Ge, V.K.Lindroos, *J. Phys. IV (France)*, **112**, 1009 (2003).
10. N.Glavatska, *J. Ferroelectrics*, **V290-292**, 93 (2003).
11. N.Glavatska, I.Glavatskiy, *Material Sci. Forum, ECRS6*, **404-407**, 841 (2002).
12. N.I.Glavatska, A.A.Rudenko, V.A.L'vov, *J. Magn. Magn. Mater.*, **241**, 287 (2002).
13. V.A.L'vov, A.A.Rudenko, N.Glavatska, *Phys. Rev. B*, **71 024421**, 1 (2005).
14. V.T.Cherepin, N.I.Glavatska, I.N.Glavatskiy, V.G.Gavriljuk, *Measur. Scien. and Techn.*, **13**, 174 (2001).
15. N.Glavatska, G.Mogilniy, S.Danilkin, D.Hohlwein. in: *Mater. Sci. Forum, European Powder Diffraction EPDIC-8*, **V443-444**, 2003, p.397.
16. N.Glavatska, I.Glavatskiy, G.Mogylny et al., *J. Phys. IV (France)*, **112**, 963 (2003).
17. G.Mogylnyy, I.Glavatskiy, N.Glavatska et al., *Scripta Mater.*, **48/10**, 1427 (2003).
18. R.C.O'Handley, D.I.Paul, M.Marioni et al., *J. Phys IV (France)*, **112**, 973 (2003).
19. A.A.Likhachev, K.Ullakko, ar. Xiv:cond-mat/0005425, 24 May 2000, 3 (2000).
20. E.V.Gomonay, V.A.L'vov, *Met. Phys. Adv. Techn.*, **20**, 24 (1998).
21. V.A.Chernenko, V.A.L'vov, E.Cesari, *J. Magn. Magn. Mater.*, **196-197**, 859 (1999).
22. V.A.Chernenko, V.A.L'vov, P.Mullner et al., *Phys. Rev. B*, **67**, 064407 (2003).
23. N.I.Glavatska, A.A.Rudenko, I.N.Glavatskiy, V.A.L'vov, *JMagn. Magn. Mater.*, **265**, 142 (2003).
24. M.R.Leadbetter, G.Lindgren, H.Rootsen, *Extreme and Related Properties of Random Sequences and Processes*, N.-Y., Springer (1983).
25. N.Glavatska, G.Mogylny, I.Glavatskiy, V.Gavriljuk, *Scripta Mater.*, **V46**, 605 (2002).
26. L.Hirsinger, C.Lexcellent, *J. Magn. Magn. Mater.*, **254-255**, 275 (2003).
27. O.Heczko, N.Glavatska, V.Gavriljuk, K.Ullakko, *Mater. Sci. Forum*, **373-376**, 341 (2001).
28. L.Hirsinger, C.Lexcellent, *J. Magn. Magn. Mater.*, **254-255**, 275 (2003).
29. O.Heczko, N.Glavatska, V.Gavriljuk, K.Ullakko, *Mater. Sci. Forum*, **373-376**, 341 (2001).

## **Залежні від часу ефекти, спричинені дією магнітного поля у Ni–Mn–Ga мартенситах з магнітною пам'яттю форми**

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Стаття являє собою огляд опублікованих та нових результатів з дослідження явища деформації, спричиненої магнітним полем фіксованої величини з плином часу при сталій температурі (магнітний крип). Досліджено часову еволюцію мартенситної структури у незмінному магнітному полі, часову залежність індукованих магнітним полем деформацій та намагніченості у Ni–Mn–Ga мартенситах з магнітною пам'яттю форми. Показана аналогія у часовій еволюції деформацій під дією сталого магнітного поля та механічних напружень фіксованої величини. Досліджено вплив температури, величини та кристалографічного напрямку магнітного поля. Проаналізовано фізичні процеси, які визначають часову еволюцію мартенситної структури та, відповідно, деформації у стаціонарних умовах у фіксованому магнітному полі при сталій температурі. Запропоновано механізми часової еволюції мартенситної структури та деформації під дією сталого магнітного поля. Базуючись на теорії випадкових процесів показано, що термічні флуктуації мікронапружень, які викликає магнітне поле, відповідають за магніто-механічну поведінку Ni–Mn–Ga мартенситів з магнітною пам'яттю форми, що відбувається із плином часу. Активація нових систем двійникування у мартенситі з п'ятишаровою модульованою структурою та їх часова еволюція визначає довготривалу еволюцію мартенситу у стаціонарних умовах у разі, коли напрямок магнітного поля співпадає з "жорсткою" віссю намагніченості.