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Fluctuating stress as the origin of the time-dependent magnetostrain effect in Ni-Mn-Ga martensites

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A recently observed phenomenon of the time-dependent deformation of ferromagnetic Ni-Mn-Ga martensites under a stationary magnetic field has been studied. A statistical model describing this phenomenon has been deduced from the mathematical theory of random processes. The influence of the duration of measurement on the shape of strain-field curves has been observed experimentally and interpreted in the statistical model framework of the time-dependent deformation. The evolution of deformation of the Ni-Mn-Ga alloy in the stepwise magnetic field has been considered. Good agreement between the theoretical and experimental results has been achieved for the suitable set of the model parameters.

I. INTRODUCTION

The magnetostrain effect (MSE) in ferromagnetic martensites has been intensively studied and a strong deformation $\varepsilon \geqslant 5\%$ of the martensitic alloys under the action of the relatively low $(0.2-1.0$ T) magnetic field has been observed (see Refs. 1–4, and references therein). Giant magnetically controlled strain is caused by the magnetically induced motion of martensite twin boundaries (Refs. 1–7, and references therein). The magnetic 90° domain walls coincide with the twin boundaries separating the different variants of the martensitic phase.[3,4]

Since the magnetic shape memory materials are expected to have great potential for the design of a new class of magnetic actuators and sensors, the rate of dynamic response of the martensitic structure to the magnetic field action is of interest. A slow time dependence of the MSE value in the Ni-Mn-Ga magnetic shape memory alloys affected by the stationary magnetic field at a constant temperature has been recently found and studied experimentally.[8–10,4] The first attempt of a theoretical interpretation of this effect was made in Ref. 11. As a result, the following physical picture of time-dependent MSE in the martensites was substantiated.

(i) A magnetic field application results in an internal magnetomechanical 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 magnetomechanical 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.

A statistical model of the quick magnetomechanical response of martensite was elaborated on in Ref. 12. Strictly speaking, this model corresponds to the case when the microstress immediately follows the magnetic field variation and the time effects are not considered in Ref. 12. An extension of the model to the case of the slow evolution of magnetic-field-induced deformations is carried out below. The conception of a slow magnetomechanical response 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.

II. BASIC ASSUMPTIONS AND MODEL EQUATIONS

In accordance with the current views[1–11] and basing on our previous work[12] we assume the following.

(a) A martensitic structure can be modeled by the alternating spatial domains of x and y variants of tetragonal lattice, the neighboring domains form the twins.

(b) The displacements of the twin boundaries are caused by the difference in stress components $\sigma = \sigma_{xx} - \sigma_{yy}$ on the right and left of the twin boundary.

(c) The displacements have a jumplike character due to the presence of pinning forces (potential wells for the twin boundaries) in a martensitic alloy specimen.

(d) The jumps of different twin boundaries occur at different stress values and result in a gradual stress relaxation and disappearance of one of the martensite variants (twin components) forming the xy structure.

For the sake of certainty, let the stationary magnetic field **H** be applied in the y direction to the twinned Ni-Mn-Ga alloy with $c/a < 1$ (a and c are the parameters of tetragonal lattice). In this case the field application is equivalent to the compression of the x variant of martensite in the y direction without stressing the y variant. It means that $\sigma = \sigma(H) > 0$ for the x variant and $\sigma = 0$ for the y variant (for more detail see Ref. 12).

According to point (iv), the stress $\sigma(H,t)$ induced by the stationary magnetic field can be modeled by the random function $\xi(t)$ fluctuating around a certain value of $\sigma(H)$, i.e.,

$$\sigma(H,t)|_{H=\text{const}} = \xi(t), \quad \langle \xi(t) \rangle = \sigma(H). \qquad (1)$$

As is consistent with points (c) and (d), the fluctuating stress results in the jumplike displacements of the certain twin boundaries in the martensite when the total stress exceeds certain critical values σ_n (the subscript $n = 1, 2, \ldots, N_0$ enumerates the pinning forces acting on the different twin boundaries). If the positive steady stress is large enough, the negative values of the random function are improbable, hence, σ_n are positive and may be enumerated in the ascending order, i.e., $\sigma_{n-1} < \sigma_n, \forall n$. The probabilities of the twin boundary jumps are statistically distributed around the characteristic value σ_c, corresponding to the maximal frequency of jumps. The relevant probability distribution can be chosen in the Gauss-like form[12]

$$p_n = \frac{1}{Z} \exp\left\{ -\frac{(\sigma_n - \sigma_c)^2}{2\sigma_0^2} \right\},$$

$$Z = \sum_n \exp\left\{ -\frac{(\sigma_n - \sigma_c)^2}{2\sigma_0^2} \right\}. \qquad (2)$$

The random function $\xi(t)$ crosses the σ_n levels from time to time. Let the function $f(\xi_n, \dot{\xi}, t_n)$ be the probability density of crossing of the nth level in the moment t_n (here $\xi_n = \sigma_n$ and $\dot{\xi} = d\xi/dt$). The number N_n of the crossings of the nth level during the time period $(0, t)$ is expressed by the well-known Rice formula.[13] If the positive steady stress is large enough, the crossings with negative $\dot{\xi}$ values may be disregarded and Rice formula may be written down in the form

$$N_n = \int_0^t \int_0^{\dot{\xi}_{\text{max}}} f(\xi_n, \dot{\xi}, \tau) \dot{\xi} d\dot{\xi} d\tau, \qquad (3)$$

where the discrete variable t_n is replaced by the continuous variable τ and the point-by-point summation is replaced by integration.

For a stationary random process the random values ξ and $\dot{\xi}$ are statistically independent and can be characterized by the individual probability $f_\xi(\xi_n)$ and probability density $\tilde{f}(\dot{\xi})$. It means that for the stationary process $f(\xi_n, \dot{\xi}) = f_\xi(\xi_n)\tilde{f}(\dot{\xi})$. In this case the Eq. (3) gives $N_n/t = \nu f_\xi(\xi_n)$, where

$$\nu = \langle \dot{\xi} \rangle = \int_0^{\dot{\xi}_{\text{max}}} \tilde{f}(\dot{\xi}) \dot{\xi} d\dot{\xi} \qquad (4)$$

is an average rate of $\xi(t)$ variation. An average time needed for the first crossing of the nth level is expressed as

$$\langle t_n \rangle = \begin{cases} t_0, & \text{if } \sigma_n \le \sigma(H), \\ t_0 + [\nu f_\xi(\xi_n)]^{-1}, & \sigma_n > \sigma(H). \end{cases} \qquad (5)$$

(Here t_0 is the moment of magnetic field application.) Physically Eq. (5) means that the average field-induced stress $\sigma(H)$ is sufficiently large to overcome the pinning forces with the numbers from 1 to i immediately after the magnetic field application. The number $n = i$ can be found from the inequalities

$$\sigma_i < \sigma(H) \le \sigma_{i+1}. \qquad (6)$$

The jumps of the boundaries pinned by the forces with the numbers $n > i$ are caused by the fluctuating stress. These jumps are responsible for the slow evolution of deformation value in a stationary magnetic field.

In the moment $t = \langle t_n \rangle$ the fluctuating stress crosses the nth level and the total stress exceeds the critical stress value σ_n. In this moment the critical values σ_j with the numbers $j < n$, are already exceeded once or more and, therefore, the number of the twin boundaries jumped during the period of time from $t = t_0$ to $t = \langle t_n \rangle$ is expressed by the formula

$$N(n) = N_0 \sum_j p_j \theta(\sigma_n - \sigma_j), \qquad (7)$$

where θ is a stepwise Heaviside function $N_0 \equiv n_{\text{max}}$.

Further consideration of the problem is similar to that carried out in Ref. 12. According to this work the volume fraction of the y variant of martensite depends on the number n as

$$\alpha_y(n) = \alpha_y(0) + [\alpha_{\text{max}} - \alpha_y(0)]N(n)/N_0, \qquad (8)$$

where α_{max} is the maximal volume fraction of the y variant of martensite in the specimen (this value may be different than the unit due to the presence of xz twins or the residual austenitic domains in the experimental specimen). The alloy deformation in the transversal (with respect to magnetic field vector) direction satisfies the equation[12]

$$\varepsilon(n) = \frac{1}{2S}\sigma(H) + (1 - c/a)[\alpha_y(n) - \alpha_y(0)], \qquad (9)$$

where S is a longitudinal elastic stiffness of tetragonal lattice.

Equations (5) and (9) define an implicitly assigned function $\varepsilon(t)$ within the time interval $t_0 \le t \le \langle t_{N_0} \rangle$. This function can be computed numerically for the different forms of statistical distribution $f_\xi(\xi_n)$. The distribution providing the best fit of the theoretical $\varepsilon(t)$ dependencies to the experimental values measured in the stationary magnetic field can be found and, in such a way, the information about the fluctuating stress and relevant physical characteristics of the martensitic alloy can be obtained, in principle. However, as a matter of fact, the fulfillment of this scientific program is impossible now because of the indefiniteness in the values of the model parameters. Nevertheless, the theoretical studies checking the statistical model itself may be carried out preliminary and the values of the model parameters providing a reasonable agreement between the theoretical and experimental data can be estimated tentatively. To this end the most frequently encountered Gauss distribution

$$f_\xi(\xi) = \frac{1}{\xi_0 \sqrt{2\pi}} \exp\left\{ -\frac{[\xi - \sigma(H)]^2}{2\xi_0^2} \right\} \qquad (10)$$

may be used for the computation of time-dependent magnetically induced deformation.

III. RESULTS

The theoretical model formulated above involves a sufficiently large number of physical parameters: the values N_0,

TABLE I. Polynomial coefficients.

η	1	2	3
B_η	-7×10^{-4}	1.019	-20.07
η	4	5	6
B_η	238.4	-755.3	1130
η	7	8	9
B_η	-882.0	340.6	-49.40

FIG. 1. Field-induced stress computed from the Eq. (11) (line) in comparison with the empirical values obtained in Ref. 17 (circles).

α_{max}, and $\alpha_y(0)$, characterizing the twin structure of martensite, the lattice parameters of martensitic phase, function $\sigma(H)$, and statistical parameters, which may be subdivided into two types. The parameters of the first type (σ_c, σ_0) are related to the pinning forces (potential wells) capturing the twin boundaries, while the parameters of the second type (ξ_0, ν) characterize the intensity and the rate of variation of fluctuating stress. The lattice parameters of martensite can be determined quite well from the x-ray and neutron-diffraction data (see, e.g., Refs. 14 and 15). The function $\sigma(H)$ can be taken from the magnetoelastic theory of MSE (Ref. 16) or obtained empirically from the results of compression-decompression cycles performed for the number of stationary values of magnetic field.[17] An accurate treatment of these results also makes an evaluation of the parameters σ_c and σ_0 possible.

The values listed above were successfully determined by different authors for different alloy compositions, experimental specimens, and temperatures. Unfortunately, the complete set of the parameters was never determined for the same specimen, and therefore, the following theoretical description of the time-dependent MSE, has, in essence, an illustrative character.

The computations are carried out using the value $c/a \approx 0.94$ and the empirical dependence $\sigma(H)$, which was determined in Ref. 17. In the field range below the field of magnetic saturation H_S this dependence was approximated by the polynomial

$$\sigma(H) = \sum_{\eta=0}^{8} B_\eta (H/H_s)^\eta. \qquad (11)$$

The coefficients of the polynomial providing a good fit to the empirical dependence are shown in the Table I, the adjusted function $\sigma(H)$ is plotted in Fig. 1 together with the empirical values. The physical mechanism responsible for these values is immaterial for the subject of the paper and is discussed elsewhere (see, e.g., Refs. 7 and 17).

The function (11) was used for computations because the empirical dependence markedly deflects from the quadratic one[16] in the range of magnetic fields $H > 0.4$ T. It should be noted that the coefficients listed in the Table I are not the essential model parameters and any analytic function providing the good fit to the empirical stress values can be used instead of Eq. (11).

As long as the dimensions of the experimental specimen are of the order of 1 cm, an average rate of the random stress
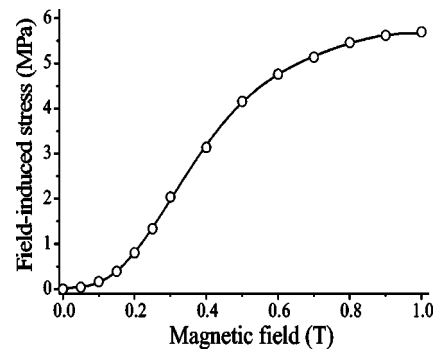
variation is evaluated by the order of magnitude as $\nu \sim \nu_t \xi_0$, where ν_t is the velocity of transverse sound wave, which, in its turn, can be estimated from the ultrasound measurements. For the quasistoichiometric Ni-Mn-Ga alloys the measurements performed in the temperature range of martensitic transformation[18] give $\nu_t \approx 3 \times 10^5$ s^{-1}. The parameters σ_c and σ_0 are sensitive to the alloy composition and mode of treatment. For the alloy specimens exhibiting almost complete reorientation of martensite variants in the saturating magnetic field these values were estimated in Refs. 12 and 17 as $\sigma_c \approx 2-4$ MPa, $\sigma_0 \approx 1$ MPa, but the largest values may be expected for the specimens exhibiting only a partial MSE. The value ξ_0 characterizes the intensity of thermal fluctuations of microstresses and will be estimated below.

Figure 2 shows the theoretical (solid line) and experimental (circles) time dependencies of deformations induced by the stepwise magnetic field. The theoretical magnetic-field-induced strains are computed for the suitable set of the model parameters presented in Table II; the line connecting the discrete theoretical values $\varepsilon(n)$ is smoothed by the "spline" tool. The parameter α_{max} was adjusted to obtain the correct maximal value of deformation observed in the experiment. The adjusted value is less than unit, and therefore, the field-induced martensitic transformation in the given alloy specimen is not complete. In this connection the values of the parameters σ_c and σ_0 presented in the Table II are somewhat larger then those reported in Refs. 12 and 17.
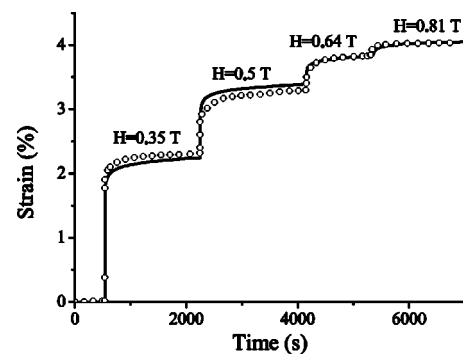

FIG. 2. Time-dependent MSE in the stepwise magnetic field: theory (curves) and experiment (circles). The values of the model parameters used for computations are shown in Table II.

TABLE II. The model parameters.

N_0	$\alpha_y(0)$	α_{max}
300	0	0.75
H_S (T)	S (GPa)	σ_c (MPa)
1	1.1	4.7
σ_0 (MPa)	ξ_0 (MPa)	ν (GPa s^{-1})
2.6	0.4	40

As long as the theoretical curve is close to the experimental one, it is worth while to analyze the model results in more detail and compare them with the available experimental data. Figure 3 illustrates the time dependence of deformation computed for the case when the stationary field $H=0.35$ T was applied at $t=0$ to the specimen with $\alpha_{max}=1$ (the values of the other model parameters are equal to those presented in the Table II). It may be concluded from the vertical segment of Fig. 3(a) that the quick magnetomechanical response of the specimen to the field application is characterized by the deformation of about of 1.5–1.75 %. But Fig. 3(b) shows that, in fact, the main part of this deformation is induced by thermal fluctuations of stress during the time period $\Delta t \gg 1/\nu_t$, while the quick magnetomechanical response, i.e., deformation induced by the average field-induced stress Eq.
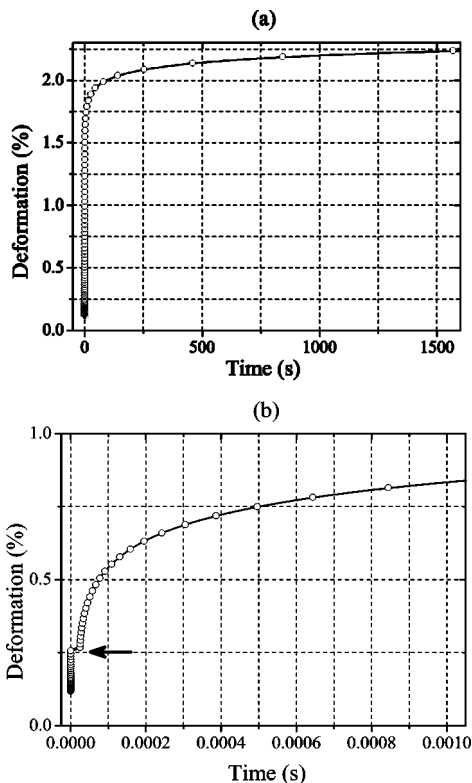

(a)

(b)

FIG. 3. Theoretical time dependence of deformation in the stationary magnetic field $H=0.35$ T. (a) and (b) show the same dependence within two different intervals of time; the needle points to the deformation value induced by the average stress $\sigma(H)=2.6$ MPa in the moment of the field application.
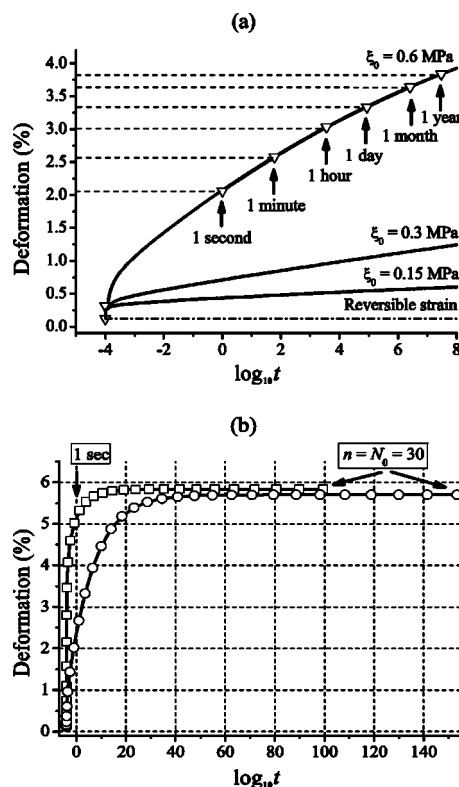

(a)

(b)

FIG. 4. Magnetic-field-induced deformation versus the common logarithm of time. (a) The curves computed for $N_0=300$, $H=0.35$ T, and different values of parameter ξ_0 characterizing the intensity of stress fluctuations. The values of the other model parameters are presented in the Table II. The down triangles mark off the standard time intervals. (b): The discrete $\varepsilon(n)$ values computed for $N_0=30$, $H=0.35$ T (circles), and $H=H_S=1$ T (squares).

(11) and conventionally attributed to the moment $t=0$, is equal to 0.25% only. In the reality the quick magnetoelastic response arises during the short time period $\Delta t \sim 1/\nu_t$ after the magnetic field application as a result of the uncontrolled transient process. Disregarding the transient process results in the existence of the small horizontal segment at the initial part of $\varepsilon(t)$ curve. The length of this segment corresponds to the time period, which is necessary for the first crossing of the critical level by the random function $\xi(t)$.

The role of stress fluctuations in the formation of magnetostrain effect is pronouncedly illustrated by Fig. 4(a). The figure shows that the reduction of the intensity of stress fluctuations results in a drastic decrease of MSE. Moreover, it is quite obvious from Fig. 4(a) that the MSE value does not reach the saturation even after one year.

The time needed for the saturation of time-dependent MSE and, accordingly, complete reorientation of martensite in a stationary magnetic field can be estimated from the Fig. 4(b). For the sake of clearness the results presented in this figure were computed for the comparatively small number of critical stress levels $N_0=30$. Figure 4(b) shows that the fluctuating stress overcomes the last critical level in the moment $t=10^{153}$ s if $H=0.35$ T and in the moment $t=10^{100}$ s if $H=1$ T. This means that the complete transformation of the twin structure into the tetragonal lattice by the magnetic field
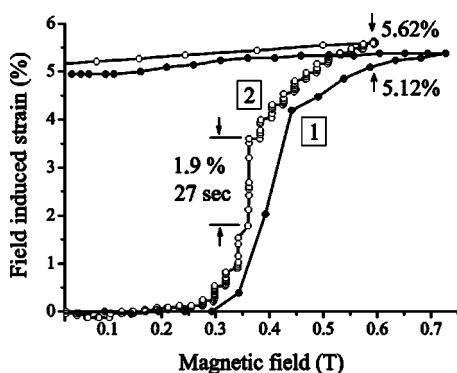
FIG. 5. Effect of the time exposure under magnetic field on the magnetostrain effect: (1) the time per step of the magnetic field change is 5 s and (2) the time per step of the magnetic field change is 27 s.

application is practically impossible if the characteristic stress σ_c is as large as 4.7 MPa. The computations show that the situation is quite different for the smaller values of characteristic stress: the deformation induced by the magnetic field $H = 1$ T reaches the saturation after the expiration of 10^3 s if $\sigma_c = 2.3$ MPa and only 1 s if $\sigma_c = 2.1$ MPa.

IV. DISCUSSION AND CONCLUSIONS

In this section the interrelation between the obtained theoretical results and available experimental data will be discussed.

Time-dependent MSE in a step-wise magnetic field. A good agreement between the experimental and theoretical values of deformations induced by the stepwise magnetic field indicates the applicability of the elaborated statistical model to the time-dependent MSE observed in Ni-Mn-Ga alloys. However, the model results can be interpreted now only qualitatively, in view of indeterminacy in the values of model parameters.

MSE in a variable magnetic field. The computations show that the time dependence of MSE is observable within both long and short time intervals (Figs. 3 and 4). As far as the values of magnetostrain induced by the variable magnetic field are always measured at appropriate intervals of time, the shapes of experimental strain—field curves must depend on the duration of the experiment. A direct experimental evidence of the validity of this statement is presented in Fig. 5 (the details of experiment are explained in the original work by Glavatska *et al.*[9]). This figure shows that the duration of experiment not only affects the value of magnetostrain but also changes the characteristic magnetic field value, making it look similar to the field of saturation of MSE. In contrast, the saturation field estimated from the magnetization curves must be independent of the duration of measurement if the field is applied in the "hard direction" z of the single-crystalline specimen (i.e., when the magnetic field does not change the martensitic structure). It can be concluded, therefore, that the field of saturation of the magnetically induced deformation may be different from the field of magnetic

saturation. Some experimental evidences supporting this conclusion can be found in the literature but its rigorous verification requires special experiments.

Temperature dependence of MSE. As was shown recently, the experimental value of MSE strongly depends on the temperature (see Refs. 4, 15, and 19–21). The saturation value of MSE is limited by the theoretical limit $(1 - c/a)$. In the single crystal exhibiting a large MSE of about 5% (and, respectively, a small σ_c value), the magnetically induced deformation rises with cooling down the specimen due to the increase of c/a ratio.[4,15,19,20] In contrast with this, the authors[21] have observed comparatively small MSE's (and rather large σ_c value) in the specimen with approximately the same c/a ratio when the temperature was close to the martensitic transformation temperature but have observed no MSE well below this temperature. Seems the twin boundaries in this specimen[21] were hardly pinned by defects, and only the intensification of thermal fluctuations arising from the temperature increase enabled the jumps of the boundaries. According to Fig. 4(a) it can be supposed that MSE in the sample[21] may be enlarged by the long-time exposure in the stationary magnetic field due to the cooperative action of the field and thermal fluctuations of microstresses.

In accordance with the results reported above, the magnetomechanical response to a fixed magnetic field must slow down with the temperature decrease. The experimental observation of a deceleration of time-dependent strain in a steady magnetic field with cooling was reported in Ref. 10.

The theoretical study of MSE in the framework of the statistical model and the comparison of theoretical and experimental data result in the following general conclusions.

(I) Thermal fluctuations of microstresses strongly affect the magnetomechanical properties of the ferromagnetic martensites typified by the Ni-Mn-Ga alloys.

(II) Thermal fluctuations of microstresses not only exhibit themselves as the time-dependent MSE observed in the protracted experiments with a stationary magnetic field, but also affect the commonly discussed experimental dependence of the deformation versus magnetic field.

(III) A well elaborated by the mathematicians theory of random processes[22] provides a powerful tool for the broad theoretical study of the time-dependent MSE and related to it phenomena. Moreover, the Rice formula (3) can be used, it seems, for the comprehension of the broad scale of nonmagnetic phenomena observed in the loaded alloys, including some aspects of creep flow, mechanical twinning, strengthening, and edging.

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