Magnetic-Field-Induced Stresses and Magnetostrain Effect in Martensite

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An equivalence principle for mechanical and magnetoelastic stresses is used for the quantitative theoretical description of giant magnetostrain effect in ferromagnetic martensite. Field-induced strains are computed in the framework of phenomenological magnetoelastic model for two different orientations of magnetic field with respect to the crystal axes. Good agreement between the theoretical and experimental field dependencies of the strains in Ni–Mn–Ga alloys is achieved.

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1. Introduction

Some alloys undergoing a cubic-tetragonal martensitic transformation (MT) are ferromagnetic. The Curie temperatures $T_C$ of these ferromagnets may be either higher or lower than the martensite start temperatures $T_{ms}$.\textsuperscript{[1–4]} The Ni–Mn–Ga and Fe–Pd ferromagnetic alloys are of a special interest because the substantial change in martensitic structure of these alloys can be induced not only by the temperature variation or mechanical stressing but also by the magnetic field application. As far as the change in alloy structure is accompanied by its deformation, the giant magnetostrain effect (GMSE) is observed in the Ni–Mn–Ga and Fe–Pd ferromagnetic martensites\textsuperscript{[5,6]} and the record-breaking field-induced strains $\varepsilon(H)$ $\sim$ 5% are achieved (see, e.g., Ref. 7).

Different approaches to the theoretical description of ferromagnetic martensite have been elaborated. Theoretical models proposed in Refs. 6, 8, 9) consider the volume fractions of martensite variants as the thermodynamical variables. These models are focused on the thermodynamical analysis of GMSE without the consideration of physical interactions originating the effect. Another approach to the theoretical study of ferromagnetic martensite is close to the traditional Landau theory and is referred to as the phenomenological magnetoelastic model.\textsuperscript{[10–14]} A spin-lattice interaction is explicitly regarded in this model by introduction of the appropriate terms into the Landau potential. In this way, the magnetic anomalies accompanying MT have been described\textsuperscript{[10,13]} and the phase diagram of ferromagnetic martensite has been constructed.\textsuperscript{11} Furthermore, a resemblance between the ordinary mechanical stressing of specimen and the stress caused by the magnetic field application has been used for the evaluation of GMSE in saturating magnetic field.\textsuperscript{[12–15]} An enlargement of magnetic-field-induced strains in the temperature range of coexistence of parent and martensitic phases was deduced from the magnetoelastic model.\textsuperscript{13} (This effect was observed experimentally, e.g., in Refs. 14, 16)). At the same time, the dependencies of the field-induced strains $\varepsilon$ on the magnetic field value $H$ have not been analyzed in framework of magnetoelastic model.

In this paper the studies initiated in Refs. 12, 13) are completed by the quantitative theoretical description of $\varepsilon(H)$ dependencies in the alloys exhibiting GMSE. The difference in experimental curves obtained in Ref. 5) for the different field orientations is explained. A quantitative agreement between the theoretical and experimental curves $\varepsilon(H)$ is achieved for the martensite in the recently studied near stoichiometric Ni$_2$MnGa alloy.\textsuperscript{14}

2. Magnetoelastic Model of Martensite

2.1 General considerations

The role of magnetoelastic coupling in the properties of martensitic alloys becomes clear when the Gibbs potential of the alloy is presented in the following form:\textsuperscript{[13]}

$$G = F_c + F_m - \delta_0 y^2 u_1 - \frac{1}{6}(\sigma_2^{me} u_2 + \sigma_3^{me} u_3),$$  \hspace{1cm} (1)

where

$$\sigma_2^{me} = \sigma_2 + \sigma_2^{me}, \quad \sigma_3^{me} = \sigma_3 + \sigma_3^{me},$$

$$\sigma_2 = \sqrt{3}(\sigma_{xx} - \sigma_{yy}), \quad \sigma_3 = 2\sigma_{zz} - \sigma_{yy} - \sigma_{xx},$$

$$\sigma_2^{me} = 6\sqrt{3}\delta_3 (m_x^2 - m_y^2), \quad \sigma_3^{me} = 6\delta_1 (2m_x^2 - m_y^2 - m_z^2),$$

where

$$u_1 = (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})/3$$ \hspace{1cm} (2)

\varepsilon_{ij} and $\sigma_{ij}$ are the components of strain and stress tensors, respectively, coordinate axes $x$, $y$ and $z$ are oriented along [100], [010] and [001] crystallographic directions respectively.

The term

$$F_c = \frac{3}{2}(C_{11} + 2C_{12})u_1^3 + \frac{1}{6}C'(u_2^2 + u_3^2),$$

describes free energy of elastic strains ($C_{11}$, $C_{12}$ and $C' = (C_{11} - C_{12})/2$ are the elastic moduli of cubic crystal).
The term \( F_m \) is a free energy of magnetic subsystem of crystal. This energy can be expressed as

\[
F_m = \frac{1}{2} J(T) y^2 + \frac{1}{2} M^2(T) (\mathbf{m} \cdot \mathbf{D} \cdot \mathbf{m}) - \mathbf{m} \mathbf{H} M(T),
\]

where the first summand is the spin exchange energy, the second summand is the magnetostatic energy caused by magnetic dipole-dipole interaction and the third one is the energy of magnetization vector \( \mathbf{M} \) in the magnetic field \( \mathbf{H} \). All energies are expressed through of the dimensionless variables \( \hat{m} = M(T)/M(T) \) and \( y = M(T)/M(0) \).

The term with parameter \( \delta \) in the eq. (1) describes the volume magnetostriction. The functions \( \sigma_{\text{eff}} \) and \( \sigma_{\text{el}} \) can be interpreted as the “effective stresses". \(^{12, 13} \) These stresses are the sums of the ordinary mechanical stresses \( \sigma_2, \sigma_3 \) and magnetoelastic ones \( \sigma_{\text{me}}^{\ast}, \sigma_{\text{eff}}^{\ast} \). The latter are caused by magnetoelastic interaction and therefore they are proportional to the magnetoelastic parameter \( \delta \) responsible for the magnetostrictive shear deformation of crystal lattice. Magnetoelastic stresses depend on the direction of unit magnetic vector \( \mathbf{m} \) and hence, a rotation of the magnetic vector under the action of magnetic field results in additional stressing of the alloy. The field-induced stresses are

\[
\sigma_{2,3}(H) = \sigma_{2,3}^{\ast}(H) - \sigma_{2,3}^{\ast}(0).
\]

For the parent phase the equilibrium conditions \( \partial(F_0 + F_{\text{me}})/\partial u_{2,3} = 0 \) result in the implicit relationships \( u_{2,3}(H) = \sigma_{2,3}(H)/2C \) between the strains and applied magnetic field. For the spatially inhomogeneous martensitic phase the theoretical strain-field dependencies can be derived phenomenologically with regard to the similarity between the mechanical and field-induced stresses. Indeed, the potential eq. (1) comprises mechanical and magnetoelastic stress tensors in combination \( \hat{\sigma}_{\text{eff}} = \hat{\sigma} + \hat{\sigma}_{\text{me}}(0) + \hat{\sigma}(H) \), and therefore, the field-induced deformation is equal to the deformation caused by mechanical load, if the field-induced stress tensor \( \hat{\sigma}(H) \) is equal to the stress tensor \( \hat{\sigma} \) caused by the loading of experimental specimen. This conclusion may be referred to as the equivalence principle for the mechanical and magnetoelastic stresses.

The structural changes accompanying mechanical stressing of shape-memory alloys cause a superelasticity and, in some cases, a rubber-like behavior of these alloys. In these cases the large strain is induced by the small mechanical stress, hence, the stiffness coefficient relating the applied mechanical stress to the resultant deformation of alloy is substantially smaller, than \( C' \). Thus, for the initial part of \( \varepsilon - \sigma \) curve the relationship \( \varepsilon(\sigma) \approx (\partial \varepsilon/\partial \sigma)_{\text{el}} \cdot \sigma \) is acceptable, where \( (\partial \varepsilon/\partial \sigma)_{\text{el}} \gg 1/C' \). In accordance with the equivalence principle we can assert that the field-induced deformation can be expressed similarly, i.e.

\[
\hat{\varepsilon}(H) \approx (\partial \hat{\varepsilon}/\partial \sigma_{\text{me}}) \cdot \sigma_{\text{me}}(H),
\]

where the coefficients \( (\partial \hat{\varepsilon}/\partial \sigma_{\text{me}}) \) are comparable in value with the derivatives \( (\partial \varepsilon/\partial \sigma)_{\text{el}} \) obtained from the ordinary mechanical tests of alloy. It can be concluded, that the giant magnetoelastic response of ferromagnetic martensite essentially is a superelastic and/or rubber-like behavior of alloy caused by the field-induced stresses. This conclusion explains a substantial variance in the experimental values of magnetoelastic response reported in literature for the different alloys of Ni–Mn–Ga system: the initial slopes and the general forms of \( \sigma - \varepsilon \) curves measured for these alloys strongly depend on their temperature, composition and preliminary treatment.

Tetragonal lattice with the principal symmetry axis directed closely to \([100], [010], \) or \([001]\) direction of parent phase may be referred to as the \( x-, y- \) or \( z\)-variant of tetragonal phase correspondingly. The spontaneous strains emerged as a result of tetragonal distortion of cubic phase can be characterized by the parameter \( u_0 = 2(c_1 - a_1)/a_1 \), where \( a_1 \) and \( c_1 \) are the parameters of tetragonal unit cell. The periodic microstructure formed by the alternating variants of low-temperature phase is typical for the large number of martensites. In the case when the period of martensitic microstructure is substantially smaller than the dimensions of ferromagnetic domains, the magnetic vector \( \mathbf{m} \) of every domain is coupled with the average strains rather than with the strains inherent to the individual martensite variants. This case was analyzed in Refs. 10, 12 and an agreement between the theoretical results and experimental data obtained in Refs. 1, 3 was achieved. Meanwhile, the very small magnetic domains and comparatively large elastic twins of tetragonal phase were observed recently in some Ni–Mn–Ga alloys. \(^{17, 18} \) As far as a twinning period exceeds in this case the dimensions of magnetic domains, the magnetic field application results in the rotation of magnetic vectors inside the tetragonal martensite variants.

In the case when the field-induced deformations expressed by eq. (5) are substantially smaller than the spontaneous strains, accompanying MT, the field dependence of magnetoelastic stresses and strains can be found by the further development of the phenomenological theory of ferromagnetic martensite. \(^{10} \) According to this theory, the magnetic anisotropy energy of \( j \)-variant of tetragonal phase can be expressed as

\[
F^{(j)} = -a M^2(T) m_3^2,
\]

where the anisotropy constant \( a = 3A_{\text{an}} \) is positive for the “easy axis" type of magnetic anisotropy being peculiar to Ni–Mn–Ga alloys with \( T_m < T_C ; \delta = \delta_1/M^2(T) \) is a dimensionless magnetoelastic constant.

The energy eq. (6) being proportional to \((1 - c_1/a_1)\) describes the magnetic anisotropy of tetragonal phase, while the magnetoelastic energy involved in the Gibbs potential eq. (1) corresponds to the cubic symmetry of parent phase. It can be shown, that the symmetry lowering of magnetoelastic interaction is described by the energy terms proportional to \((1 - c_1/a_1)^2 \). As far as these terms are disregarded, the present theory corresponds to the linear approximation in small parameter \((1 - c_1/a_1)\).

Further consideration is carried out for the ellipsoidal specimen with the axes aligned with \([\bar{1}10], [\bar{1}10] \) and \([001]\) directions. In appropriate coordinate system a demagnetization matrix is diagonal, its elements are denoted as \( D_1, D_2, D_3 \), respectively. For every variant of martensite the angle between the magnetic vector \( \mathbf{m} \) and field direction can be determined from the minimum conditions for the energy \( F = F_m + F^{(j)} \) being the function of polar coordinates \( \varphi, \psi \). The vector \( \mathbf{m} \) components are

\[
m_x = \sin \psi \cos \varphi, \quad m_y = \sin \psi \sin \varphi, \quad m_z = \cos \psi.
\]
2.2 Strains induced by the field applied along [001]

Field applied along [001] || z rotates the magnetic vectors $\mathbf{m}_1$ and $\mathbf{m}_2$ of x- and y-variants in planes $\varphi_1 = \text{const}$ and $\varphi_2 = \pi/2 - \varphi_1$, respectively. In this case

$$\cos \psi = \cos \psi_{[001]} = \begin{cases} H/H_{S1}, & H < H_{S1}, \\ 1, & H \geq H_{S1}, \end{cases}$$

where

$$H_{S1} = M(T)(D + 2(a \cos^2 \varphi_1 - D_{xy} \sin \varphi_1 \cos \varphi_1)),$$

$$D' = D_1 - (D_1 + D_2)/2, \quad D_{xy} = (D_1 - D_2)/2,$$

$$\cos^2 \varphi_1 = \frac{1}{2} + \frac{a}{2\sqrt{a^2 + D_{xy}^2}},$$

$$\sin \varphi_1 \cos \varphi_1 = -\frac{D_{xy}}{2\sqrt{a^2 + D_{xy}^2}}.$$

The field value $H = H_{S1}$ corresponds to magnetic saturation of x- and y-variants. The magnetic vector of z-variant is aligned with applied magnetic field.

As far as $m_{1z}^2 - m_{2z}^2 = \sin^2 \psi \cos 2\varphi$, the $\sigma_2(H)$ stress components induced in x- and y-variants have the same absolute value but differ in sign and therefore, compensate each other in average. At the same time, for both x- and y-variants of martensite the eq. (4) and the definition of $\sigma_{zz}^c$ values result in the formula $\sigma_2(H) = 18\delta M^2(T) \cos^2 \psi_{[001]}$. For z-variant the field-induced stresses are absent in view of steady vector $\mathbf{m}$ direction, and therefore the stress component $\sigma_3(H)$ should be multiplied by factor 2/3. Thus, the experimentally measured giant magnetoelastic response is caused by stress component $\sigma_3(H)$ and the formula for macroscopic deformation is

$$\langle \hat{\varepsilon}(H, T) \rangle \approx \frac{2}{3} \cdot (\partial \hat{\varepsilon}/\partial \sigma_3) \cdot 18\delta M^2(T) \cos^2 \psi_{[001]}.$$ (8)

Equation (8) predicts the strong temperature dependence of field-induced deformation of martensite in the temperature ranges close to $T_m$, $T_c$ or intermartensitic transformations temperatures, because $\partial \hat{\varepsilon}/\partial \sigma_3$ and/or $M$ values critically depend on the temperature in these ranges. A possibility of GMSE in the two-phase state existing in the vicinity of martensite start temperature was deduced theoretically in Ref. 13 and confirmed experimentally (see, e.g., Refs. 14, 16).

2.3 Strains induced by the field applied along [110]

When the magnetic field is oriented in [110] direction the magnetic vectors $\mathbf{m}_1$ and $\mathbf{m}_2$ of x- and y-variants are directed in such a way that

$$m_{1z} = m_{2z} = 0, \quad m_{1x}^2 = m_{2x}^2, \quad m_{1y}^2 = m_{2y}^2,$$

for any magnetic field value (Fig. 1). In this case the field-induced stress component $\sigma_3(H)$ is equal to zero for both variants, while the $\sigma_2(H)$ component inherent in x-variant differs in sign from one applied to y-variant, and therefore, these components compensate each other. Thus, the x- and y-variants do not contribute to macroscopic deformation. The field rotates the magnetic vector $\mathbf{m}_3$ of z-variant in plane $\varphi = \pi/4$. For the specimen with $D_2 = D_3$ the equations for angle $\psi$ are

$$\sin \psi = \sin \psi_{[110]} = \begin{cases} H/H_{S2}, & H < H_{S2}, \\ 1, & H \geq H_{S2}, \end{cases}$$

where $H_{S2} = M(T)(2a + D_1 - D_3)$. As far as $m_{3x} = m_{3y} = \text{const}$, the component $\sigma_2(H)$ is equal to zero. At the same time, the magnetic field induces a nonzero stress component $\sigma_3(H) = -18\delta M^2 \sin^2 \psi_{[110]}$ in z-variant of martensite. Thus, the macroscopic deformation induced by the field applied in [110] crystallographic direction is

$$\langle \hat{\varepsilon}(H, T) \rangle \approx -\frac{1}{3} \cdot (\partial \hat{\varepsilon}/\partial \sigma_3) \cdot 18\delta M^2 \sin^2 \psi_{[110]}.$$ (10)

(The factor 1/3 appears in eq. (10) because only one of the martensite variants contributes to the macroscopic deformation).

A comparison of eqs. (8) and (10) shows, that the strain tensor components induced by the saturation field $H = H_{S2}$ applied in [110] direction differ by the factor $-1/2$ from the appropriate components induced by the field $H = H_{S1}$ applied along [001]. The difference in the saturation field values is caused by the magnetostatic energy, and therefore, depends on the shape of experimental specimen. The magnetostatic energy is considered here in a traditional way, which, exactly speaking, should be modified with account of the distinctive features of martensitic state. One of the possible approaches to this difficult problem is developed in, but its detailed consideration is out of the scope of the present paper.

3. Computations

The computation of field-induced deformation is carried out with regard to an imperfection of crystal lattice (see Refs. 14, 19). The imperfection of the martensitic structure results in the random dispersion of $u_1$ variable. The “statistical” value of this variable is $u_i^{(0)} = u_i^{(0)} + s$, where $u_i^{(0)}$ characterizes the volume change accompanying the cubic-tetragonal phase transformation, $s$ denotes the dispersion. The term $-\delta_0 y^2 u_1$ in eq. (1) gives rise to the renormalization of exchange parameter $J^*$ introduced in eq. (3): $J(T) \rightarrow$
The field-induced deformation is $J^*(T, s) = J(T) - 2\delta_0 t^{(s)}$. Therefore, the Curie temperature found from the condition $J^*(T, s) = 0$ is also randomly dispersed:

$$T_C(s) = T_C + 2s\frac{T_C\delta_0}{\xi},$$

where $\xi$ is the constant in the relationship $J^*(T, 0) = \xi(T - T_C)/T_C$ postulated in the Landau theory. Physically $T_C(s)$ values are the temperatures of ferromagnetic ordering in the small spatial domains of the imperfect experimental specimen. For these domains the magnetization value may be found from well-known equations

$$M(T, s) = M(0)y(T, s), \quad y(T, s) = \tanh[T_C(s)y(T, s)/T].$$

(12)

Now it should be noted that the eqs. (8) and (10) for the field-induced deformation also depend on the random value $s$ through the $M$, $\psi_{[001]}$, and $\psi_{[110]}$ values. When the volume fractions of $x$-, $y$-, and $z$-variants are equal to each other the average field-induced deformation is

$$\langle \hat{\varepsilon}(H, T) \rangle = 12\delta(\delta_0/\sigma_\Sigma)$$

$$\cdot \left[ \int_{-\infty}^{+\infty} M(T, s) \cos \psi_{[001]}f(s)\theta(T_C(s) - T)ds \right]^2,$$

(13)

when the magnetic field is aligned with [001], and

$$\langle \hat{\varepsilon}(H, T) \rangle = -68(\delta_0/\sigma_\Sigma)$$

$$\cdot \left[ \int_{-\infty}^{+\infty} M(T, s) \sin \psi_{[110]}f(s)\theta(T_C(s) - T)ds \right]^2,$$

(14)

when the field is parallel to [110] direction. Here $f(s) = \frac{1}{s_0\sqrt{2\pi}} \exp \left( -\frac{s^2}{2s_0^2} \right)$ is the Gauss distribution, $\theta(T_C(s) - T)$ is the step-wise Heaviside function. (More details concerning the magnetization process in martensite and the averaging procedure are reported in Refs. 10, 11, 19).

Equations (13), (14) are used for the computation of the magnetic-field-induced strains in Ni–Mn–Ga martensitic specimen studied in Ref. 5). This specimen, is as a platelet oriented normally to [110] direction. Such platelet is modelled by the ellipsoid flattened in [110] direction (see Fig. 1). In such a case $D_1 > D_2 = D_3$. The $D_1/D_2$ ratio is found from magnetization curves and $(\delta\varepsilon_{[001]}/\sigma_\Sigma)$ value is considered as fitting parameter. All other parameters involved in the computations are taken from Refs. 5, 10, 12, 14). In particular, the value $\delta = -23$ is accepted for the dimensionless magnetoelastic constant. This parameter is obtained from the value $\delta_1 = -1.2$ MPa reported in Refs. 10, 11, 12) for the Ni–Mn–Ga alloy with $M(T_{m}) \approx 7 \cdot 10^5$ JT$^{-1}$m$^{-3}$.

Figure 2 shows the field-induced deformation of the specimen along [001] axis computed for the field applied in [001] and [110] directions. A semi-quantitative agreement between the theoretical and experimental dependencies $\varepsilon_{[001]}(H)$ is achieved for $(\delta\varepsilon_{[001]}/\sigma_\Sigma)^{-1} = 59$ MPa/percent. This value correlates with the experimental values $\sigma/d\varepsilon \sim 100$ MPa/percent experimentally obtained in the course of mechanical stressing of Ni–Mn–Ga alloys (see Ref. 20 and references therein).

An absence of the rigorous quantitative agreement between the theoretical and experimental $\varepsilon_{[001]}(H_{[001]})$ dependencies presented in Fig. 2 may be attributed to the experimental details. In support of this statement a theoretical analysis of the experimental data obtained in a very recent work (Ref. 14) for the near stoichiometric Ni$_2$MnGa alloy with $T_{m} = 160$ K is carried out. The parallelepiped-shaped specimen of this alloy with the dimensions of $5 \times 7 \times 11.5$ mm$^3$ is modelled by the ellipsoid with the axes $A = B = 6$ mm, $C = 11.5$ mm. The values $T = T_0 = 140$ K, $M(T_0) = 5.25 \cdot 10^5$ JT$^{-1}$m$^{-3}$ are taken for computations in accordance with real experimental conditions and previously accepted value $\delta = -23$ is used.

Figure 3 illustrates a good quantitative agreement between the theoretical and experimental $\varepsilon_{[001]}(H)$ curves achieved for...
Fig. 4 The influence of the statistical dispersion of Curie temperature on the field dependence of deformation. The solid line computed for $s_0 = 0$, $(\partial \varepsilon_{001}/\partial \sigma_3)^{-1} = 37$ MPa/percent demonstrates parabolic dependence while the bold line obtained for $s_0 = 0.030$, $(\partial \varepsilon_{001}/\partial \sigma_3)^{-1} = 41$ MPa/percent exhibits the deviation from the parabola.

4. Conclusions

The following conclusions about the physical nature of GMSE in the martensites can be made on the basis of the equivalence principle for the mechanical and magnetoelastic stresses formulated above.

1) The GMSE observed in some martensitic alloys is a manifestation of the ordinary spin-lattice interactions (existing in all ferromagnetic solids) in combination with the superelasticity or rubber-like behavior of these alloys.

2) The advancement in a phenomenological magnetoelastic model of the ferromagnetic martensite enables a numerical computation of the field-induced deformation and provides the quantitative agreement between the theory and experiment (see Fig. 3).

3) In particular, the magnetoelastic model explains the difference between $\varepsilon_{001}(H_{001})$ and $\varepsilon_{001}(H_{110})$ dependencies observed experimentally in Ref. 5.

It is worth noting that the equivalence principle for the mechanical and magnetoelastic stresses enables a preliminary estimation of the expected magnetoelastic response of an alloy from the precisely measured stress–strain curves.

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