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Low Temperature Physics 48, 206–211 (2022)

<https://doi.org/10.1063/10.0009538>



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Landau theory of ferroelastic phase transitions: Application to martensitic phase transformations

Cite as: Fiz. Nizk. Temp. 48, 231–237 (March 2022); doi: 10.1063/1.0009538

Submitted: 21 January 2022



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ABSTRACT

The peculiarities of two Landau-type theories of ferroelastic phase transitions have been analyzed. The predictions of both theories have been compared with well-known experimental data obtained for the shape memory alloys (SMAs) undergoing the first-order martensitic transformations (MTs). It has been shown that the predictions of the Landau–Devonshire theory, which disregards the third-order term in power expansion of Gibbs free energy, contradict the experimental data, while the results of the symmetry conforming Landau theory, which takes into account this term, are in agreement with experimental data. The impossibility of occurrence of the second-order MT is demonstrated starting from the thermodynamic definition of the second-order phase transition. It is argued that the stress–strain loops, obtained for SMAs, can imitate the stress-induced phase transition in the absence of such transition.

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1. INTRODUCTION

Landau theory of phase transitions is widely used for the description of physical effects accompanying the ferroelastic phase transitions in multiferroic materials. A ferroelastic phase transition is defined in the Landau theory as the phase transition characterized by the order parameter composed of the strain tensor components. The equilibrium value of the order parameter is equal to zero in the high-symmetry (parent) phase and has nonzero value in the low-symmetry (product) phase.

The Landau theory of ferroelastic phase transitions is applicable to the solids exhibiting the appearance of spontaneous strain on cooling below the phase transition temperature. Two different variants of the Landau theory of phase transitions are used for the description of these phase transitions. The first one is a modification of Landau–Devonshire (LD) theory advanced long ago for *ferroelectric* phase transitions; it is based on the power expansion of Gibbs free energy, which involves only even powers of the order parameter.^{1,2} This expression is referred to as the 2–4–6 Gibbs free energy, where the numbers show that this expression involves the second-, fourth-, and sixth-order terms in the order parameter of ferroelastic phase transition.³ The second variant of the theory is based on the 2–3–4 series expansion of Gibbs free energy that is the expression,

which involves the second-, third-, and fourth-order terms in the order parameter.^{4–7}

The martensitic transformations (MTs) of shape memory alloys (SMAs) are the widely studied phase transitions of ferroelastic type (see, e.g., handbook⁸ and references therein) because these alloys are extensively applied in the engineering and medicine. Therefore, they appeared to be good candidates for the application of the Landau theory of ferroelastic phase transitions.

In the present communication, we emphasize that the predictions of LD theory contradict to the well-known experimental data obtained for MTs in SMAs, while the conclusions drawn from the theory, which takes into account the third-order term in power expansion of Gibbs free energy, are in agreement with these data. We emphasize that the adequate theory of ferroelastic phase transition must be symmetry conforming, i.e., the tensor character of the order parameter must be taken into account.

2. LANDAU–DEVONSHIRE THEORY APPLIED TO FERROELASTIC PHASE TRANSITIONS

The LD theory^{9,10} was advanced first for ferroelectric phase transitions, which are characterized by the spontaneous (arising in absence of external electric field) electric polarization of solid. The LD theory was based on the minimization of Gibbs free energy

expressed through the components of polarization vector \mathbf{P} as

$$G = \frac{1}{2}\chi^X(P_x^2 + P_y^2 + P_z^2) + \frac{1}{4}\xi^X(P_x^4 + P_y^4 + P_z^4) + \frac{1}{2}\lambda(P_y^2P_z^2 + P_z^2P_x^2 + P_x^2P_y^2) + \frac{1}{6}\zeta^X(P_x^6 + P_y^6 + P_z^6) \dots \quad (1)$$

It should be stressed, first, that Eq. (1) is invariant with respect to the reversal of sign of polarization vector components. Due to this, the electrically polarized states with $\mathbf{P} = \mathbf{P}_0$ and $\mathbf{P} = -\mathbf{P}_0$ have the same energy. Therefore, the LD theory shows that the spatial domains of electrically polarized state with the opposite directions of vector \mathbf{P} have to arise on the cooling of solid below the phase transition temperature. This theoretical conclusion is in agreement with the great number of experiments (see, e.g., Ref. 10 and references therein). It should be noticed, second, that the minimization of Gibbs free energy leads to conclusion that the spontaneous electric polarization of solid is a first-order phase transition if coefficient $\xi^X < 0$ and a second-order one if $\xi^X > 0$. Negative and positive values of this coefficient are reported for BaTiO₃, PbTiO₃, SrBi₂Nb₂O₉ and SrTiO₃, LiTaO₃, LiNbO₃, Sr_{0.8}Bi_{2.2}Ta₂O₉, respectively.¹⁰ The first-order ferroelectric phase transitions are observed in BaTiO₃, 0.32PIN–0.345PMN–0.335PT,¹¹ while the phase transformations showing the features of second-order transitions, are observed in SrTiO₃, CuInP₂Se₆.^{12,13} Therefore, the LD theory is adequate to ferroelectric phase transitions.

The ferroelastic phase transition is the transformation of cubic crystal lattice into the crystal lattice possessing the tetragonal, orthorhombic, rhombohedral, or monoclinic symmetry. To demonstrate that the LD theory, which starts from 2–4–6 potential, is not adequate to ferroelastic phase transitions, let us consider the application of this theory to cubic-tetragonal MT in SMA, see, e.g., Ref. 14. This MT results in the appearance of non-zero values of the diagonal strain tensor components and is characterized by the “tetragonality” of crystal lattice, $1 - c/a$, where $a = b$ and c are the lattice parameters measured in the tetragonal phase. In accordance with the basic principle of Landau theory of phase transitions the power series expansion of Gibbs free energy must include all terms, which are invariant with respect of the symmetry group of the parent phase. The group theory shows the existence of the third-order invariants composed from the multicomponent order parameters of ferroelastic phase transitions. However, the application of LD theory to the martensitic phase transitions (see, e.g., Refs. 15–20) ignores the third-order terms and starts from the 2–4–6 Gibbs free energy expressed as

$$G = G_0(T) + \frac{1}{2}\alpha_1\varepsilon^2 + \frac{1}{4}\alpha_2\varepsilon^4 + \frac{1}{6}\alpha_3\varepsilon^6 - \sigma\varepsilon, \quad (2)$$

where ε is relative elongation of the crystal in [001] crystallographic direction, σ is mechanical stress, $\alpha_{1,2,3}$ are phenomenological constants.⁸ The point is that the *tensor* character of the order parameter was ignored and third-order term was omitted, as it was done in the case of *vector* \mathbf{P} . The third-order term must be taken into account, and that is why the analog of potential (1) cannot be used for the description of ferroelastic phase transitions.

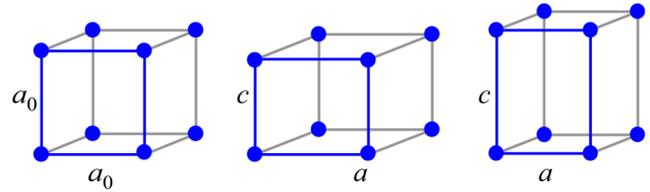


FIG. 1. The unit cell of cubic (parent) phase, and tetragonal phases with $c < a$, and $c > a$.

In essence, Eq. (2) is similar to Eq. (1), because both equations express the Gibbs free energy of 2–4–6 type. Due to this, the theory based on Eq. (2) is referred to as the LD model of MTs.^{1,2} This model pretends to the description of the transformational properties of SMAs. It should be emphasized, however, that the predictions of the theory, which is based on Eq. (2), drastically contradict experimental data. This theory predicts that

- (i) The mixed two-phase state formed by the tetragonal phases with $1 - c_1/a_1 > 0$ and $1 - c_2/a_2 = -(1 - c_1/a_1) < 0$ must appear as the result of MT in the unstressed specimen because these phases have the same energy. (The Gibbs free energy Eq. (2) in the absence of mechanical stress is invariant with respect to the change of variables $\varepsilon \rightarrow -\varepsilon$.)
- (ii) The MT must be the second-order phase transition if $\alpha_2 > 0$ and the first-order phase transition if $\alpha_2 < 0$.

However, the numerous experiments show that

- (A) The MTs never result in the mixed states formed by the spatial domains of tetragonal lattices with the opposite signs of tetragonality. The formation of such states does not happen, because the crystal cells with $1 - c/a < 0$ are not equivalent to the crystal cells with $1 - c/a > 0$ (see Fig. 1), and therefore, the crystal lattices formed by such cells have different energy values.
- (B) The second-order MTs are not observed. (Only the quasi second-order MTs were realized in some non-stoichiometric alloys by the precise tuning of their chemical composition, aimed to minimization of tetragonality of product phase.^{21,22})

The disagreement between the theory based on Eq. (2) and experimental data is caused by the fact that the transformation of strain tensor components ε_{ik} under the operations of the symmetry group is similar to this of bilinear combinations of polarization vector components P_iP_k .²³ Therefore, 2–4–6 expression for Gibbs free energy, is transformationally equivalent to 1–2–3 expression composed of strain tensor components.

3. SYMMETRY CONFORMING LANDAU THEORY FOR FERROELASTIC PHASE TRANSITION

3.1. Symmetry conforming equation for Gibbs free energy

For the ferroelastic phase transitions, the order parameter components are the linear combinations of the strain tensor components. To provide the minimum for the Gibbs free energy, the second-, third-, and fourth-order invariant terms must be included

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in the power series for Gibbs free energy. The first-order terms describe the heat expansion of solids, and so, they can be disregarded by the phase transition theory.

This Gibbs free energy is expressed through the two-component order parameter of cubic-tetragonal ferroelastic phase transitions as²⁴

$$G = \frac{1}{2}A_1u_1^2 + \frac{1}{2}A_2(u_2^2 + u_3^2) + \frac{1}{3}B_1u_1^3 + \frac{1}{2}B_2u_1(u_2^2 + u_3^2) + \frac{1}{3}B_3u_3(u_3^2 - 3u_2^2) + \frac{1}{4}C_1u_1^4 + \frac{1}{2}C_2u_1^2(u_2^2 + u_3^2) + \frac{1}{3}C_3u_1u_3(u_3^2 - 3u_2^2) + \frac{1}{4}C_4(u_2^2 + u_3^2)^2 - \frac{1}{6}(\sigma_2u_2 + \sigma_3u_3), \quad (3)$$

where the order parameter components are

$$u_1 = (\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz})/3, \quad u_2 = \sqrt{3}(\epsilon_{xx} - \epsilon_{yy}), \\ u_3 = 2\epsilon_{zz} - \epsilon_{xx} - \epsilon_{yy}.$$

The phenomenological constants $A_{1,2}$, B_{1-3} , C_{1-4} are the linear combinations of elastic modules (see, e.g., Ref. 25). The Gibbs free energy Eq. (3) and its simplified versions are used in the symmetry conforming theory of ferroelastic phase transitions and may be referred to as 2–3–4 model. The detailed analysis of Eq. (3) is given in Ref. 26 and in the Appendix.

3.2. Explanation of basic experimental data

It was proved long ago that the presence of third-order term makes impossible the second-order phase transition.^{26,27} This statement of phase transitions theory leads to the conclusion that all ferroelastic phase transformations are the first-order phase transitions (see point B). To explain point A the minimum conditions for the Gibbs free energy must be considered (see Appendix). Three equivalent domains with principal axes parallel to x , y , and z have the same energy. For definiteness, the domain with a principal axis along z will be considered. For this domain the minimum conditions for Gibbs potential result in the equations, which have solution $u_2 = 2$ and $u_3 = 3\epsilon_{zz}$ (see Appendix). Due to this, Eq. (3) is reduced to the form

$$G = \frac{1}{2}A_2(T)u_3^2 + \frac{1}{3}B_3u_3^3 + \frac{1}{4}C_4u_3^4 - \frac{1}{6}\sigma_3u_3, \quad (4)$$

where $C_4 > 0$, the coefficient $A_2(T)$ changes its sign during the phase transition. For the unstressed specimen the condition $\partial G/\partial u_3 = 0$ gives the equilibrium value of the order parameter

$$u_3 = 2(c/a - 1) = -(B_3/2C_4) \left(1 + \sqrt{1 - 4A_2(T)C_4/B_3^2} \right), \quad (5)$$

which corresponds to the minimum of Gibbs free energy (see, e.g., Ref. 24). The austenitic phase is stable if $A_2(T) > 0$, while the martensitic phase is stable if $A_2(T) < B_3^2/4C_4$, in the temperature interval $0 < A_2(T) < B_3^2/4C_4$ these two phases coexist. The coexistence of two phases is the basic feature of first-order phase transition. The equations $A_2(T) = 0$ and $A_2(T) = B_3^2/4C_4$ prescribe the lability

temperatures of austenitic and martensitic phases, respectively. The phase transition results in the stabilization on tetragonal lattice with $c < a$, if $B_3 > 0$, or $c > a$, if $B_3 < 0$. The sign of B_3 is fixed if the phase transition in the certain solid is considered. Due to the presence of cubic term in Eq. (4) the energies of tetragonal phases with $c < a$ and $c > a$ are different at all temperatures and therefore *these phases do not coexist*. This result of 2–3–4 potential explains point A that is in agreement with plenty of experiments performed for MT. The description of MTs in the stressed specimens is presented in Ref. 28.

3.3. Imitation of quasi second-order phase transition by shape memory alloys

Equation (5) describes the temperature-induced ferroelastic phase transition.²⁹ However, Eqs. (3) and (4) are often used for the description of the stress-induced phase transitions observed in the course of stress-strain cycles.³⁰ These equations made it possible

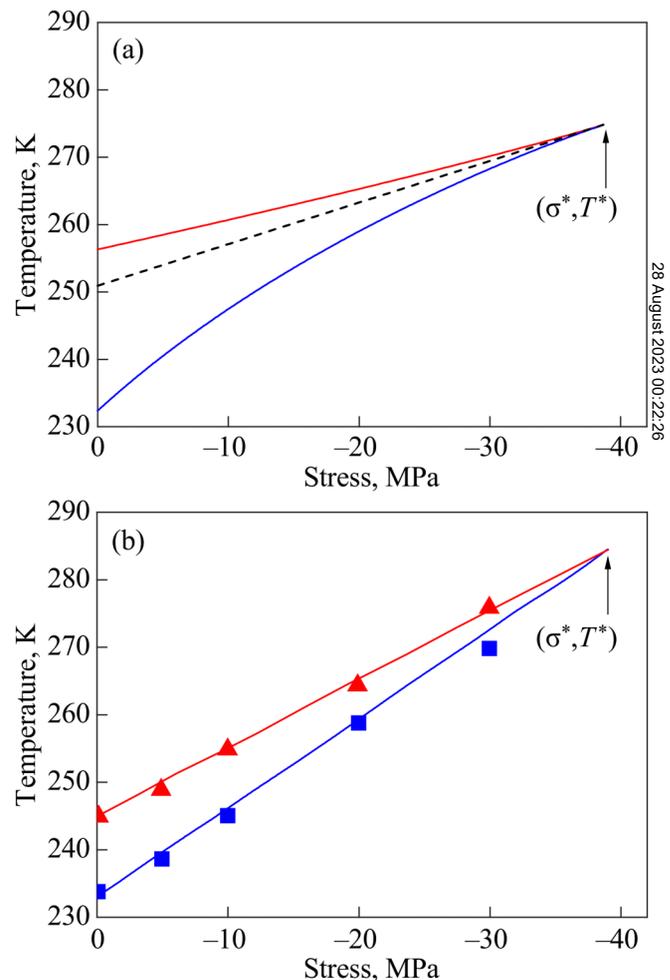


FIG. 2. The theoretical (a), and experimental (b) phase diagram of MT obtained for Fe–Pd alloy.^{31,32}

to obtain the stress–temperature phase diagrams for SMAs. A theoretical phase diagram computed for Fe–Pd alloy is shown in Fig. 2(a).³¹ This diagram consists of lability lines of martensitic and austenitic phases, the area between these lines corresponds to the mixed austenite–martensite state. The dashed line is the phase transition line. At this line, the energy of austenitic phase is equal to the energy of martensitic phase. It is worth noting, that the phase transition line is not an average line between austenite and martensite lability lines, but lies closer to the martensite lability line. The lability lines meet in the critical point (σ^*, T^*). This is the end-point of the phase transitions line. Therefore, the stress–strain cycles performed at the temperature exceeding critical temperature T^* are not accompanied by the stress-induced MTs. Experimental data confirming the presence of critical point are shown in Fig. 2(b).³²

In spite of the conclusion of symmetry conforming phase transition theory, the transformational behavior, which may be attributed to the second-order phase transition, was observed for some axially stressed SMAs.^{33–36} The theoretical explanation and prediction of the results of stress–strain tests performed above the critical temperature T^* attract a special attention of researchers dealing with SMAs, because the realization of giant anhysteretic deformation is promising for various applications. However, the fundamental problem arises in the course of traditional interpretation of experimental results. To explain the essence of this problem, the existence of the end-point (σ^*, T^*) of the first-order phase transition line at the stress–temperature phase diagram must be taken into account. Figure 3 shows the theoretical stress–strain loops computed in Ref. 28 for the temperatures $T < T^*$ and $T > T^*$ using the 2–3–4 power expansion for Gibbs free energy and taking into account the presence of crystal defects in the deformed SMA. The figure illustrates drastic difference in transformational behavior of an alloy above and below critical temperature: the bottom loop

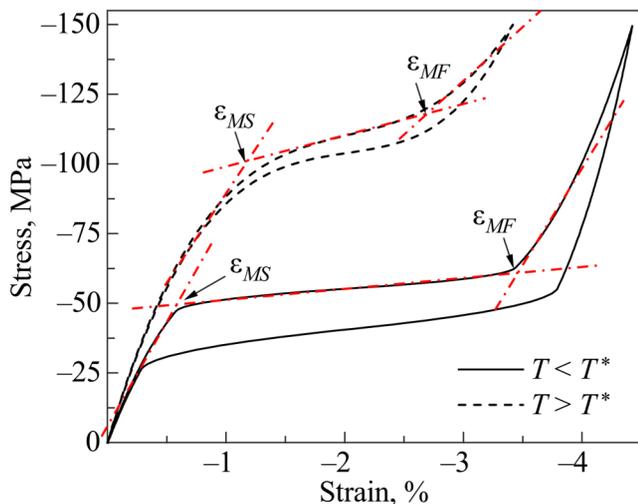


FIG. 3. The stress–strain loops computed in the presence (solid line) and in the absence (dashed line) of stress-induced phase transition.²⁸

shows pronounced hysteresis with plateau-like segments, meanwhile the upper loop shows gradual strain increase with much smaller hysteresis.

The pronounced hysteresis is caused by the first-order phase transition, the much smaller hysteresis is caused by the crystal defects and takes place in the absence of phase transition. It is important that the absence of phase transition cannot be noticed if the standard procedure is used for determination of the strain values corresponding to start (ϵ_{MS}) and finish (ϵ_{MF}) of MT. This procedure attributes the start and finish of the stress-induced MT to the deflection points from the linear segments of the stress–strain loops (see straight dash-dotted lines in Fig. 3) and to the existence of hysteresis. However, despite of the commonly accepted

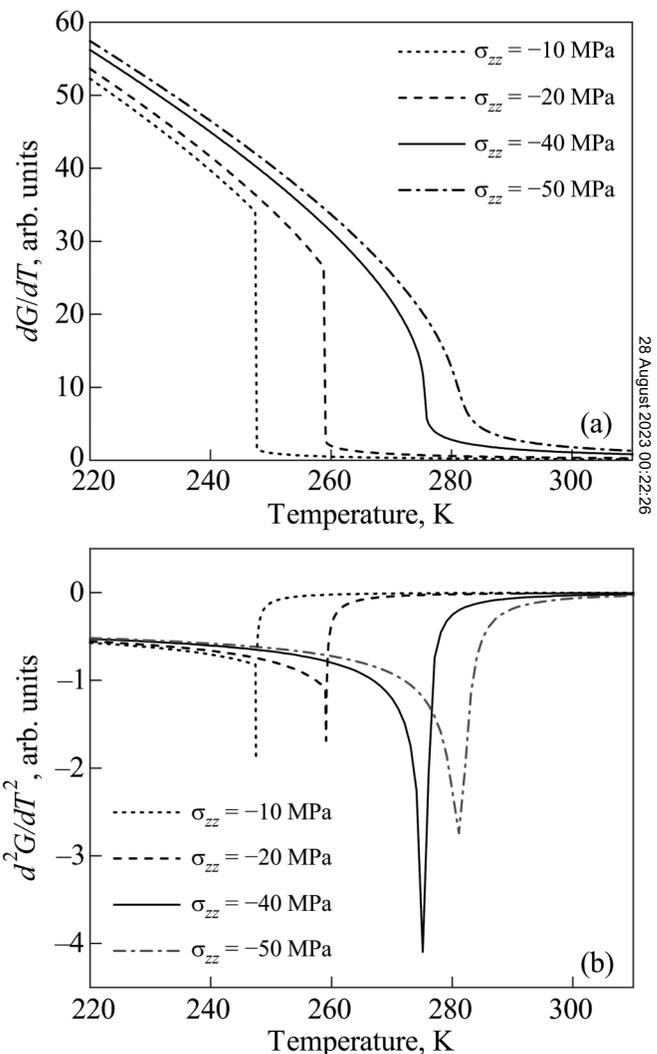


FIG. 4. The first (a), and second (b) derivatives of Gibbs potential on temperature computed for different values of applied mechanical stress.

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interpretation, the upper loop doesn't exhibit the martensitic transformation. This loop characterizes the gradual deformation of crystal lattice, without jumps of strain value. The large non-linear deformations of SMAs in the absence of phase transition, predicted in theoretical work,²⁸ were observed experimentally then for Ni-Fe-Co-Ga and Fe-Pd alloys.^{37,32} However, there are still disputes concerning the possibility of observation of second-order MT in SMAs, as so this statement must be supported by the fundamental thermodynamic arguments.

According to thermodynamic definition, the first-order phase transition is the change of thermodynamic state characterized by the jump-like change of the first derivative of thermodynamic potential, while the second-order phase transition must be accompanied by the jump-like change of the second derivative of thermodynamic potential. Figures 4(a) and 4(b) show the first and second temperature derivatives of Gibbs potential, respectively. These values were computed using Eq. (4) for Fe-Pd alloy, the model parameters used for computations were estimated previously (see Ref. 31) from the experimental phase diagram.³² Fig. 4(a) shows the decrease of the jump of first derivative of Gibbs potential with increasing applied mechanical stress. When applied stress is equal to critical stress $\sigma^* = -40$ MPa (see phase diagram Fig. 2) the jump of first derivative vanishes indicating the end of first-order MT. This is in agreement with experimental data,³² which point to the disappearance of first-order MT at critical stress -40 MPa. The Fig. 4(b) shows that the second derivative of Gibbs potential varies continuously if the mechanical stress is larger than critical stress $\sigma^* = -40$ MPa, and therefore, neither first-order phase transition nor second-order phase transition can be observed on cooling/heating of alloy specimen.

4. CONCLUSION

The consistent theoretical analysis of Landau-type theories showed that

- (1) The predictions of 2-4-6 theory, which disregards the third-order summand in the power expansion of Gibbs potential in terms of strain tensor components, contradict the well-known experimental data obtained for MTs in SMAs and could not be used for the description of MTs. In the same time, the symmetry conforming theory, which takes into account the third-order term in power expansion of Gibbs free energy, is applicable to MTs and is in agreement with experimental data.
- (2) The standard procedure of determination of stress-induced MT from experimental stress-strain loops, can result in the wrong conclusion about MT if the alloy temperature is slightly higher than the critical temperature, corresponding to the end-point of phase transition line in the stress-temperature phase diagram. The reason is that the non-linearity of stress-strain curves and small hysteresis of stress-strain loop may be observed even in the absence of MT.
- (3) Computed temperature dependences of first and second derivatives of Gibbs potential are in agreement with thermodynamic definitions of the first-order and the second-order phase transition. These computations explicitly prove the absence of temperature-induced phase transition in the stressed alloy

when the stress exceeds the critical value corresponding to the end-point of phase transition line.

ACKNOWLEDGMENTS

The authors are grateful to Professor V. A. L'vov for fruitful discussions. This work was performed under the financial support of National Research Foundation of Ukraine (Grant No. 2020.02/0261). A.K. acknowledges the scholarship of Verkhovna Rada of Ukraine for young scientists (Grant No. 0121U112445).

APPENDIX: MINIMIZATION OF GIBBS POTENTIAL

The equation for the Gibbs potential, which takes into account both order parameter components $u_{2,3}$ and concomitant component u_1 , can be written in the form:

$$G = \frac{1}{2}A_1u_1^2 + \frac{1}{2}A_2(u_2^2 + u_3^2) + \frac{1}{2}B_2u_1(u_2^2 + u_3^2) + \frac{1}{3}B_3u_3(u_3^2 - 3u_2^2) + \frac{1}{3}C_3u_1u_3(u_3^2 - 3u_2^2) + \frac{1}{4}C_4(u_2^2 + u_3^2)^2 - \frac{1}{6}(\sigma_2u_2 + \sigma_3u_3). \quad (A1)$$

The minimum conditions for this Gibbs free energy are $\partial G/\partial u_1 = 0$, $\partial G/\partial u_2 = 0$, $\partial G/\partial u_3 = 0$. The first condition gives

$$u_1 = -[B_2(u_2^2 + u_3^2) + C_4u_3(u_3^2 - 3u_2^2)]/2A_1.$$

The u_1 is proportional to the second power of order parameter components and therefore, $u_1^2 \ll u_2^2 + u_3^2$. This inequality corresponds to the volume conservation principle, which is inherent to the phase transitions observed in the martensitic alloys. (The strong inequality shown above explains why the terms proportional to u_1^3 , u_1^4 are omitted in Eq. (A1).) In the case, if the mechanical load is applied in the [001] crystallographic direction the value $\sigma_2 = 0$ and Gibbs potential is equal to

$$G = \frac{1}{2}A_2(u_2^2 + u_3^2) + \frac{1}{3}B_3u_3(u_3^2 - 3u_2^2) + \frac{1}{4}C_4(u_2^2 + u_3^2)^2 - \frac{1}{6}\sigma_3u_3. \quad (A2)$$

The minimum conditions with respect to order parameter components are

$$\begin{aligned} \partial G/\partial u_3 &= A_2u_3 + B_3u_3^2 + C_4u_3^3 - \frac{1}{6}\sigma_3 = 0, \\ \partial G/\partial u_2 &= A_2u_2 - 2B_3u_3u_2 + C_4u_2^3 = 0. \end{aligned} \quad (A3)$$

The second equation in (A3) results in the equality $u_2 = 0$. As so the Eq. (A3) can be simplified to the Eq. (4) in the cases of unloaded specimen and specimen mechanically loaded in the z direction. The Eq. (4) is used also for the accounting of the influence of temperature dependence of lattice parameters on the ferromagnetic resonance in martensitic films.³⁸

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It can be concluded that the minimization of Gibbs potential with respect to both components of two-component order parameter reduces Eq. (3) for Gibbs potential to Eq. (4) and it is an exact result of the 2–3–4 theory.

It should be noted that the volume conservation principle is not a general feature of ferroelastic phase transitions in the solids. Phase transitions with pronounced volume change are observed in particular in Mn–As and Fe–Rh alloys. The power expansion of Gibbs potential with respect to u_1 variable³⁹ and magnetoelastic model⁴⁰ are used for the description of phase transitions in Mn–As and Fe–Rh, respectively.

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