## EQUATIONS OF STATE DETERMINING THE MAGNETOELASTIC PROPERTIES OF FERROMAGNETIC MONOCRYSTALS

K. B. VLASOV

Institute of Metal Physics, Academy of Sciences, U.S.S.R.

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A thermodynamic derivation is given of the equations of state (for small magnetization changes and deformations) that determine the magnetomechanical properties of magnetically polarized ferromagnetic monocrystals possessing hexagonal symmetry. It is shown that in the case of nonuniform strains, the equilibrium values of the mechanical stresses and of the magnetic field intensity are connected by equations of state not only with the magnetization vector and strain tensor, but also with the rotation tensor, which determines the orientation of the volume element under consideration. On the basis of the equations obtained, conclusions are drawn regarding special features of the velocity of propagation and rotation of the plane of polarization of transverse elastic waves in ferromagnets.

1. In works of the author,  $^{1,2}$  equations of "state" have been obtained that describe the magnetoelastic properties of magnetically polarized magnetoelastic media in the dynamic regime; and on the basis of these equations, it has been shown<sup>3</sup> that in the propagation of transverse elastic waves along the direction of the magnetization, a rotation of their plane of polarization is to be expected. To determine the physical nature of certain constants that enter in the equations mentioned above and in the equation for the angle of rotation of the plane of polarization, it is necessary to consider specific types of magnetoelastic media.

2. We consider ferromagnetic monocrystals. To make the calculation more definite, we choose for consideration ferromagnetic monocrystals possessing hexagonal symmetry, in which the sixfold axis is the axis of easy magnetization. With respect to their elastic, magnetic, and magnetoelastic properties for small magnetization changes, such monocrystals behave like magnetoelastic magnetically polarized media possessing uniaxial symmetry, whose properties were treated in references 1-3. We shall furthermore consider equilibrium processes and treat the crystal as infinite.

Let a change of the magnetization I occur in such a monocrystal, and let the monocrystal undergo an arbitrary elastic deformation, defined by the vector displacement u of an element of volume as a whole, the strain tensor  $\epsilon_{ij} = \frac{1}{2} (\partial u_i / \partial x_j + \partial u_j / \partial x_i)$ , and the rotation tensor  $\omega_{ij} = \frac{1}{2} (\partial u_i / \partial x_j - \partial u_j / \partial x_i)$ of the volume element. The equations of state that connect the equilibrium values of the strains and magnetization with the magnetic field intensity **H** and with the symmetric components  $\sigma_{ij}$  and the antisymmetric components  $\sigma_{ij}^*$  of the mechanical "stress" tensor  $\sigma_{ij}^{**}$  can be found from the condition that the thermodynamic potential

$$\Phi = u_{\mathbf{ex}} + u_{\mathbf{c}} + u_{\mathbf{me}} + u_{\mathbf{el}} - H_m I_m - \sigma_{ij} z_{ij} - \sigma_{ij}^* \omega_{ij}, \quad (1)$$

be a minimum; here  $u_{ex}$ ,  $u_c$ ,  $u_{me}$ , and  $u_{el}$  are respectively the exchange, magnetocrystallineanisotropy, magnetoelastic (magnetostrictive), and elastic energies. They are defined as follows:

$$u_{\text{ex}} = \frac{1}{2} a_1 |\mathbf{I}|^2 + \frac{1}{4} a_2 |\mathbf{I}|^4, \tag{2}$$

$$u_{\rm me} = \frac{1}{2} q_{mnij} I_m I_n \varepsilon_{ij} , \qquad (3)$$

$$u_{\mathbf{e}\mathbf{l}} = \frac{1}{2} C_{ijkl} \varepsilon_{ij} \varepsilon_{kl}. \tag{4}$$

For the case of monocrystals of hexagonal symmetry, when the  $x_3$  coordinate axis is chosen along the sixfold axis, the magnetocrystalline-anisotropy energy in the first approximation is equal to

$$u_{c} = K_{1} |[\mathbf{I} \times \mathbf{k}]|^{2} / |I|^{2},$$
(5)

where  $\mathbf{k}$  is the unit vector along the  $x_3$  axis. In this case the tensor  $q_{mnij}$  has the following independent nonvanishing components:

$$q_{1111} = q_{2222}, \quad q_{3333}, \quad q_{1122} = q_{2211}, \quad q_{1133} = q_{2233},$$

$$q_{3311} = q_{3\,322}, \quad q_{2323} = q_{2332} = q_{3223} = q_{3232} = q_{1313} = q_{1331}$$

$$= q_{3113} = q_{3131}, \quad q_{1212} = q_{1221} = q_{2112} = q_{2121}$$

$$= (q_{1111} - q_{1122}) / 2.$$
(6)

The choice of  $u_{me}$  in the form (3), with (6) taken into account, assumes that in magnetically uniaxial monocrystals, the magnetostriction is not determined solely by the orientation of the vector magnetization, as had been assumed earlier,<sup>4,5</sup> but depends also on the value of the magnitude of the magnetization.<sup>6</sup>

The tensor  $c_{ijkl}$ , in contrast to the tensor  $q_{mnij}$ , is invariant with respect to interchange of pairs of indices ij and kl. Therefore it has the same form as the tensor  $q_{mnij}$  except that  $c_{3311} = c_{1133}$ .

3. As an initial state, we may choose the state of the ferromagnetic crystal that one obtains by applying along the sixfold axis a uniform, constant polarizing magnetic field  $H_0k$ , strong enough so that the crystal will be uniformly magnetized along the  $x_3$  axis. From the minimization condition for the thermodynamic potential (1), we find the equilibrium values of the magnetization  $I_0k$  and of the stresses  $\sigma_{11}^0$  in the initial state:

$$H_{0} = (a_{1} + a_{2}I_{0}^{2})I_{0}, \qquad (7)$$
  
$$\sigma_{11}^{0} = \sigma_{22}^{0} = \frac{1}{2}q_{3311}I_{0}^{2},$$

$$\sigma_{33}^0 = \frac{1}{2} q_{3333} I_0^2, \qquad \sigma_{12}^0 = \sigma_{23}^0 = \sigma_{31}^0 = 0.$$
 (8)

In practice, it is convenient to choose another state as initial state, namely in which the initial stresses are zero. This is dependent on the fact that the crystal is usually not in an absolutely rigid environment, and under the influence of the stresses  $\sigma_{ij}^0$  it is uniformly strained, i.e., acquires uniform strains  $\epsilon_{11}^0 = \epsilon_{22}^0$  and  $\epsilon_{33}^0$ .

4. We now consider the case in which there are present in the crystal a weak magnetic field **h** and mechanical stresses, alternating both in time and in space. In the crystal there will occur deformations determined by the strain tensor  $\epsilon_{ij}$  and the rotation tensor  $\omega_{ij}$  of the volume element, and the vector magnetization in each volume element will change its value slightly both in magnitude and in direction.

Expressions (2) - (5) for the various energies have already been written in a definite coordinate system, related to the crystallographic axes [that is, a system in which the tensor constants have a completely definite form characteristic of the given crystal symmetry, for example the form (6)]; they are correct, strictly speaking, only for uniform strains and rotations (which were not contemplated in the theory of elasticity). In the general case, nonuniform strains are connected with nonuniform rotations of the volume elements of the crystal, and consequently also with nonuniform rotations of the crystallographic axes of the volume elements.

We assume that the form of the expressions (2) - (5) for the energy densities, in which the tensor constants have a completely definite form charac-

teristic of the given type of symmetry, remains valid also for nonuniform strains, but that these energies must be referred to coordinate axes  $x'_1$ ,  $x'_2$ ,  $x'_3$  related to the crystallographic axes of the volume element under consideration.<sup>7</sup> We therefore introduce two coordinate systems: 1) a fixed coordinate system  $x_1$ ,  $x_2$ ,  $x_3$  attached to the crystallographic axes of the crystal in its initial state, with the  $x_3$  axis oriented along the sixfold axis; 2) a local movable coordinate system  $x'_1$ ,  $x'_2$ ,  $x'_3$ attached to (and rotating with) the volume element under consideration, with the  $x'_3$  axis oriented along the sixfold axis of the given volume element.

5. We now obtain the explicit form of the thermodynamic potential in the fixed system of coordinates. We first write the energies  $u_c$  and  $u_{me}$ in the movable coordinate system. For this purpose it is necessary in formula (5) to replace the unit vector  $\mathbf{k}$  by the unit vector  $\mathbf{k'}$ , connected with  $\mathbf{k}$  by the relation

$$\mathbf{k}' = \mathbf{k} + [\mathbf{\omega} \times \mathbf{k}],\tag{9}$$

and in the expression (3) to substitute the values of the components of the vector magnetization and of the strain tensor in the movable coordinate system,  $I'_m$  and  $\epsilon'_{ij}$ , which are connected with the values of the components of these quantities in the fixed system of coordinates by the relations

$$\mathbf{I}' = \mathbf{I} + [\mathbf{I} \mathbf{x} \boldsymbol{\omega}], \qquad \boldsymbol{\varepsilon}_{11} = \boldsymbol{\varepsilon}_{11} + 2 (\boldsymbol{\omega}_3 \boldsymbol{\varepsilon}_{12} - \boldsymbol{\omega}_2 \boldsymbol{\varepsilon}_{13}), \dots, \\ \boldsymbol{\varepsilon}_{12} = \boldsymbol{\varepsilon}_{12} + \boldsymbol{\omega}_1 \boldsymbol{\varepsilon}_{13} - \boldsymbol{\omega}_2 \boldsymbol{\varepsilon}_{23} + \boldsymbol{\omega}_3 (\boldsymbol{\varepsilon}_{22} - \boldsymbol{\varepsilon}_{11}), \dots, \qquad (10)$$

where  $\omega$  is the vector rotation of the volume element, dual to the rotation tensor  $\omega_{ij}$ , with components given by the known relations  $\omega_1 = \omega_{32} = -\omega_{23}, \ldots$ .

We next transform, according to formulas (9) and (10), to the fixed coordinate system. Here we choose as independent variables, determining the value of the change of the vector magnetization, the value of the change of absolute value  $\Delta I$  and the vector angle of rotation  $\theta$ , which determines the rotation of the vector magnetization with respect to the fixed coordinate axes. For changes of the vector magnetization that are small in comparison with its initial value  $I_0\mathbf{k}$ , its new value can be determined from the relation

$$\mathbf{I} = (I_0 + \Delta I) \left( \mathbf{k} + [\mathbf{\theta} \times \mathbf{k}] \right) - \frac{1}{2} I_0 \left( \mathbf{\theta}^2 \mathbf{k} - (\mathbf{\theta}, \mathbf{k}) \mathbf{\theta} \right), \quad (11)$$

and we obtain

$$u_{\mathbf{c}} = K_1 \{ (\boldsymbol{\omega} - \boldsymbol{\theta})^2 - (\mathbf{k}, \ \boldsymbol{\omega} - \boldsymbol{\theta})^2 \}, \qquad (12)$$

$$u_{\overline{\mathrm{me}}} = \frac{1}{2} q_{3311} I_0^2 (\varepsilon_{11} + \varepsilon_{22}) + \frac{1}{2} q_{3333} I_0^2 \varepsilon_{33} + q_{3311} I_0 (\varepsilon_{11} + \varepsilon_{22}) \Delta I + q_{3333} I_0 \varepsilon_{33} \Delta I - 2q_{2323} I_0^2 (\varepsilon_{23} \theta_1 - \varepsilon_{13} \theta_2) + (2q_{2323} - q_{3333} + q_{3311}) I_0^2 (\varepsilon_{23} \omega_1 - \varepsilon_{13} \omega_2).$$
(13)

The exchange and elastic energies have the same form in both coordinate systems, since the first depends on the absolute value of the vector magnetization, and since the expression for the second differs by terms of the third order of small quantities when written in the fixed and in the movable systems.

On using the minimization conditions  $\partial \Phi/\partial \theta = 0$ ,  $\partial \Psi/\partial \Delta I = 0$ , and  $\partial \Phi/\partial \epsilon_{ij} = 0$  for the thermodynamic potential, and on taking account of (7) and (8), we obtain the following equations of state:

$$h_{m} = \gamma_{mn}I_{n} + h_{mf}\varepsilon_{f} + h_{mq}^{*}\omega_{q},$$
  

$$\sigma_{g} = h_{gn}^{'}I_{n} + c_{gf}\varepsilon_{f} + c_{gg}^{*}\omega_{q},$$
(14)

where now by  $I_n$  are understood the small values of the change of magnetization from its initial state, and where the new symbols  $\epsilon_f$  and  $\omega_q$  have been introduced; they are connected with the previous quantities  $\epsilon_{ij}$ ,  $\omega_m$ , and  $\omega_{ij}$  by the following scheme:  $\epsilon_1 = \epsilon_{11}, \ldots, \epsilon_6 = 2 \epsilon_{12}, \ldots, \omega_6 = -2 \omega_3$  $= 2 \omega_{12}$ . Here and hereafter we shall assume that the indices m, n, i, and j run through the values 1, 2, and 3; the indices f and g through the values 1, 2, ..., 6; and the indices p and q through the values 4, 5, and 6.

Here the following components are different from zero:

$$\gamma_{11} = \gamma_{22} = (H_0/I_0 + 2K_1/I_0^2), \qquad \gamma_{33} = (H_0/I_0 + 2a_2I_0^2), (15)$$

$$h_{31} = h_{32} = \dot{h_{12}} = \dot{h_{22}} = a_{221}I_0, \qquad h_{22} = \dot{h_{22}} = a_{222}I_0, (16)$$

$$h_{32} = h_{13} = h_{23} = q_{331110}, \quad h_{33} = h_{33} = q_{333310}, \quad (16)$$
$$h_{15} = h_{24} = h_{51} = h_{42} = q_{2323}I_0,$$

$$h_{15}^* = -h_{24}^* = K_1/I_0, \tag{17}$$

$$c_{11} = c_{22} = c_{1111}, \quad c_{33} = c_{3333}, \quad c_{12} = c_{21} = c_{1122},$$
  
 $c_{13} = c_{23} = c_{31} = c_{32} = c_{1133},$   
 $c_{44} = c_{55} = c_{2323}, \quad c_{66} = (c_{11} - c_{12})/2,$  (18)

$$\dot{c_{44}} = -\dot{c_{55}} = -(2q_{2323} - q_{3333} + q_{3311})I_0^2/4$$
  
= -(2h\_{15} - h\_{33} + h\_{31})I\_0/4. (19)

If as initial state we choose not the state with the initial stresses (8), but the state usually encountered in practice, in which these stresses are absent, then we still get equations of state of the type (14), except that the constant  $c_{44}^*$  will have the new value

$$c_{44}^* = -q_{2323}I_0^2/2 = -h_{15}I_0/2.$$
 (19')

6. On further setting  $\partial \Phi / \partial \omega_m = 0$ , we get an expression for the antisymmetric component of the mechanical "stress" tensor,  $\sigma_{ij}^*$ , to which is equivalent (dual) a certain axial vector  $T_m$ . Here

$$T_p = h_{pn}^* I_n + c_{pf}^* \varepsilon_f + c_{pq}^{**} \omega_q, \qquad (20)$$

where

$$h_{51}^{*} = -h_{42}^{*} = h_{15}^{*}, \quad c_{44}^{*} = -c_{55}^{*} = c_{44}^{*}, \quad c_{44}^{*} = c_{55}^{*} = K_{1}/2,$$
(21)

and  $T_p$  is connected with  $T_m$  and  $\sigma_{ij}^*$  by relations of the type  $T_4 = -T_1/2 = \sigma_{23}^*, \ldots$ .

We shall explain the physical meaning of the antisymmetric part of the mechanical "stress" tensor  $\sigma_{ij}^*$ . The right member of (20) was obtained by differentiating, with respect to the components of the vector angle that describes the orientation of the volume element, the magnetocrystallineanisotropy and magnetoelastic energies, which depend on interaction of the magnetization (spin system) with the crystal lattice. Therefore these derivatives should give expressions for the components of the force couples (with sign reversed),  $-T_{m}^{*}$ , exerted by the spin system on the crystal lattice of the volume element under consideration. Since in equilibrium no resultant couple should act on the volume element, this couple must be balanced by forces transmitted across the surface of the volume element from its environment. These forces can be described by the antisymmetric component  $\sigma_{ij}^*$  of the mechanical "stress" tensor  $\sigma_{ij}^{**}$ . From this it follows that the force couple  $T_m^*$  exerted by the spin system on the lattice is determined by the vector  $-T_m$  ( $T_m^* = -T_m$ ) dual to the antisymmetric component  $\sigma_{ij}^*$  of the mechanical "stress" tensor. However, the resultant force and couple (which figure in the equations of elasticity theory) on the volume element are determined by the symmetric part  $\sigma_{ii}$  of the mechanical "stress" tensor (or simply of the mechanical stress  $\sigma_{ii}$ ). This is easy to show if one considers that the force on a volume element is determined by the divergence either of the tensor  $\sigma_{ii}^{**}$  or of the tensor  $T_{ij}^*$  dual to the vector force couple  $T_m^*$ , and if one takes account of the requirement that the resultant force moment must be expressed solely in the form of a surface integral.

7. In the absence of equilibrium, the derivatives of the thermodynamic potential  $\Phi$  with respect to the components of the vector  $\theta$ , which describe the orientation of the vector magnetization, determine the components of the resultant force moment that acts on the magnetization. On taking account of this and using (14), we get for this moment the relation

$$\mathbf{T} = [\mathbf{I}_0 \times \mathbf{h}^{\mathsf{ef}}], \tag{22}$$

where  $h_m^{ef} = h_m - \bar{h}_m$ , and where  $\bar{h}_m$  is that value of the magnetic field intensity which would correspond to the given values of magnetization and strain if the process were an equilibrium process; that is, we must substitute for  $\bar{h}_m$  its value determined by formula (14). Knowing the couple that acts on the magnetization, we can construct the equation of motion of the magnetization.

8. We shall compare the relations obtained with the relations deduced earlier by a formal method. Equations (14) and (20) agree with Eqs. (7) of reference 1 and (17) of reference 2. (We remark, in this connection, that in Eqs. (7) and (17) of the articles cited,  $\epsilon_4$ ,  $\epsilon_5$ ,...,  $\omega_6$  must be replaced not by  $\epsilon_{23}$ ,  $\epsilon_{13}$ ,...,  $\omega_{12}$ , but by their doubled values.) However, the constants  $h_{15}^*$ ,  $c_{44}^*$ , and  $c_{44}^{**}$  introduced earlier by the formal method are now expressed through basic constants  $I_0$ ,  $K_1$ , and qmnif that describe the ferromagnetic.

**9.** Finally, we shall apply the relations obtained to the analysis of concrete physical processes.

In reference 3 it was shown that rotation of the plane of polarization of elastic waves may be determined, in particular, by the term containing the expression  $h_{15}$  ( $h_{15} - h_{15}^*$ ), which according to (17) can be written in the form  $h_{15}^2$  (1 - K<sub>1</sub> / I<sub>0</sub>h<sub>15</sub>). It follows that the rotation of the plane of polarization is determined not solely by the magnetostriction constant, but also by the magnetocrystalline anisotropy constant. In particular, for  $h_{15} > 0$ , the sign of the angle of rotation can depend on the ratio of the quantities  $K_1$  and  $I_0h_{15}$ , which can change with change of temperature. In order of magnitude, hmf ~  $c_{ijkl}\lambda_s/I_0$ , where  $\lambda_s$  is the saturation magnetostriction, and  $c_{ijkl} \sim 10^{12} \text{ d/cm}^2$ ; thus we find that a calculation of the term  $K_1/I_0h_{15}$  (that is, of the constant  $h_{15}^*$ ) is necessary for ferromagnetics that possess a relatively large anisotropy constant and small magnetostriction. For example, for  $K_1 \ge 10^6$ erg/cm<sup>3</sup> and  $\lambda_{\rm S} \leq 10^{-6}$  this term is greater than unity.

As a second example, we calculate the speed  $c_t$  of propagation of transverse elastic waves propagated along the sixfold axis in a magnetodielectric. On solving the equation of motion of the theory of elasticity,  $\rho \ddot{\mu}_i = \partial \sigma_{ij} / \partial x_j$ , and taking account of (14), we get

$$c_{t} = c_{t}^{0} \{1 - [h_{15}(h_{15} - h_{15}^{*}) - \gamma_{11}c_{44}^{*}]/\gamma_{11}c_{44}\}^{1/2}, \quad c_{t}^{0} = (c_{44}/\rho)^{1/2}.$$
(23)

This expression differs somewhat from the expression for  $c_t$  derived by Akhiezer, Bar'yakhtar, and Peletminskii<sup>8</sup> (on going over to the case of propagation of the wave along the symmetry axis and to low frequencies); namely, in it there are additional terms, containing the constants  $h_{15}^*$  and  $c_{44}^*$ , which take account of rotations of the volume elements. In this connection we note that the expression for  $c_t$  from reference 8, mentioned above, can be easily obtained by calculating the speed of propagation of elastic waves by the formula  $c_t$ 

=  $(c_{44}^{H}/\rho)^{1/2}$ , where  $c_{44}^{H}$  is the value of the elastic modulus measured at constant magnetic field. For quasistatic processes, there is a definite relation<sup>9</sup> between the components of the elastic modulus tensor measured at constant field,  $c_{ijkI}^{H}$ , and at constant magnetization,  $c_{ijkI} = c_{ijkI}$ . For the present case this has the form  $c_{44}^{HH} = c_{44} (1 - h_{15}^2/\gamma_{11}c_{44})$ . This difference between  $c_{44}^{HH}$  and  $c_{44}$  bears the name " $\Delta g$  -effect." Thus the difference between the relations for  $c_t$  given by Akhiezer and others<sup>8</sup> and by us [formula (23)] consists in this, that in the derivation of formula (23), along with calculation of the  $\Delta g$  -effect, which received attention in reference 8, account has been taken of the rotation of the volume elements that takes place in propagation of elastic waves transverse with respect to the polarizing magnetic field H<sub>0</sub>.

We shall assume that in the initial state the ferrodielectric is in an unstressed state. Then in accordance with (15), (17), and (19'), we get

$$c_t = c_t^0 \left[ 1 - (h_{15}^2 + \frac{1}{2}h_{15}H_0) / \gamma_{11}c_{44} \right]^{1/2}$$

Thus in this case the speed of propagation of elastic waves is determined by the  $\Delta g$ -effect alone when  $H_0 = 0$ . On increase of  $H_0$  ( $H_0 \gg 2K_1/I_0$ ),  $c_t$  tends, if we take account of (15), not to  $c_t^0$  but to the value  $c_t^0 (1 - h_{15}I_0/2c_{44})^{1/2}$ , which may be either larger or smaller than  $c_t^0$  according to the sign of the magnetostriction.

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