

THERMODYNAMIC THEORY OF FERROMAGNETIC DOMAINS

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This article presents in detail those results in ferromagnetic domain theory that have been obtained recently and have not previously appeared in monographs or reviews. A significant part of the article is devoted to consideration of basic problems of the theory. A detailed description is given of those properties of the domain structure that are independent of the properties of the model. As concrete examples, the domain structures in uniaxial and cubic ferromagnets are considered. Experimental results are cited only by way of illustration. Some generally accepted ideas have proved wrong and are subjected to criticism in this article.

CONTENTS

Introduction 555
 1. Conditions for Coexistence of Phases. Surface Tension 555
 2. Domain Structures in Ferromagnets 561
 3. Bulk Properties of Domain Structures. Theory of the Ideal Magnetization (Polarization) Curve 566
 4. Theory of Fine Ferromagnetic Particles 569
 5. Theory of Nuclei of Reverse Magnetization. 570
 Appendix. Thermodynamic Theory of Magnetostriction. The Stress Tensor 572
 References 573

INTRODUCTION

It is well known that in ferromagnets there can occur the formation of a domain structure: that is, a splitting of the specimen into regions of coexisting phases with different directions of the magnetic moment M (see, for example, [1-3]). An analogous phenomenon occurs in ferroelectric materials [4,5] and also in superconductors (the intermediate state) [1,6]. Recently the existence of a domain structure has been detected in nonferromagnetic metals, under the conditions that produce the de Haas-van Alphen effect, [7,8] and in antiferromagnets. [9]

The various problems of the theory of domain structure have been treated in a large number of books and review articles. In particular, for ferromagnets there is the very popular review by Kittel [10] (see also [11,12]). The present article presents in detail those results that have been obtained recently and that have not previously appeared in monographs or reviews. A significant part of the article is devoted to consideration of basic problems of the theory. A detailed description is given of those properties of the domain structure that are independent of the properties of the model. As concrete examples, the domain structures in uniaxial and cubic ferromagnets are considered. Experimental results are cited only by way of illustration.

Some generally accepted ideas have proved to be wrong and are subjected to criticism in this article. Such criticism is necessary because the erroneous ideas mentioned have been widely disseminated and are expounded in many textbooks.

The number of papers devoted to the theory of domain structures is very large. In order not to complicate the reading, we shall restrict ourselves to the minimum number of citations; therefore the literature list will

not contain a complete bibliography. Where it was possible, we have tried to cite those books and articles of foreign authors that have been translated into Russian.

1. CONDITIONS FOR COEXISTENCE OF PHASES. SURFACE TENSION

a) **Thermodynamic relations.** In Chapter 1 we shall neglect the effects of electrostriction and magnetostriction and shall therefore make no distinction between the free energy and the thermodynamic potential. The thermodynamic relations written below for dielectrics and magnets are ones that we shall use often in what follows.

Polarization of dielectrics can be produced by two methods: by assigning either the distribution of external charges $\rho(\mathbf{x})$ or the conductor potentials φ_i . In the first case, the free energy is a quantity \mathcal{F} defined as follows:

$$\delta \mathcal{F} = (4\pi)^{-1} \int \mathbf{E}(\mathbf{x}) \delta \mathbf{D}(\mathbf{x}) d^3 \mathbf{x} = \int \varphi \delta \rho(\mathbf{x}) d^3 \mathbf{x}, \quad (1.1)$$

where \mathbf{E} is the electric field, \mathbf{D} is the induction, φ is the electrostatic potential ($\mathbf{E} = -\nabla \varphi$), and the integration is carried out over all space, including the region outside the body. It is convenient to introduce also the so-called total free energy of the body, \mathcal{F}_{tot} :

$$\mathcal{F}_{\text{tot}} = \mathcal{F} - \int [\mathbf{E}_0^2(\mathbf{x})/8\pi] d^3 \mathbf{x}, \quad (1.2)$$

$$\delta \mathcal{F}_{\text{tot}} = - \int \mathbf{P}(\mathbf{x}) \delta \mathbf{E}_0(\mathbf{x}) dV, \quad (1.3)$$

where $\mathbf{E}_0(\mathbf{x})$ is the field produced by the assigned charges $\rho(\mathbf{x})$ in a vacuum, and where $\mathbf{P}(\mathbf{x})$ is the dipole moment of unit volume. The integration in formula (1.3) extends only over the volume V occupied by the dielectric ($\mathbf{P}(\mathbf{x}) \neq 0$). If the conductor potentials φ_i are assigned, then the free energy is the quantity

$$\tilde{\mathcal{F}} = \mathcal{F} - \int (\mathbf{E}\mathbf{D}/4\pi) d^3x = \mathcal{F} - \sum_i e_i \varphi_i, \quad (1.4)$$

$$\delta \tilde{\mathcal{F}} = -(4\pi)^{-1} \int \mathbf{D}(\mathbf{x}) \delta \mathbf{E}(\mathbf{x}) d^3x = - \sum_i e_i \delta \varphi_i; \quad (1.5)$$

here e_i are charges on the conductors.

Analogous relations are valid also for magnets. If the conduction currents $\mathbf{j}(\mathbf{x})$ are assigned, and if they produce a magnetic field $\mathbf{H}(\mathbf{x})$, the free energy is defined as follows:

$$\delta \tilde{\mathcal{F}} = -(4\pi)^{-1} \int \mathbf{B}(\mathbf{x}) \delta \mathbf{H}(\mathbf{x}) d^3x = -c^{-1} \int \mathbf{A}(\mathbf{x}) \delta \mathbf{j}(\mathbf{x}) d^3x, \quad (1.6)$$

where $\mathbf{B} = \text{curl } \mathbf{A}$ is the magnetic induction, and where c is the velocity of light.

The total free energy $\tilde{\mathcal{F}}_t$ is

$$\tilde{\mathcal{F}}_{\text{tot}} = \tilde{\mathcal{F}} + \int [\mathbf{H}'_0(\mathbf{x})/8\pi] d^3x, \quad (1.7)$$

$$\delta \tilde{\mathcal{F}}_{\text{tot}} = - \int \mathbf{M}(\mathbf{x}) \delta \mathbf{H}'_0(\mathbf{x}) dV; \quad (1.8)$$

here $\mathbf{H}'_0(\mathbf{x})$ is the field produced by the assigned currents $\mathbf{j}(\mathbf{x})$ in a vacuum, and $\mathbf{M}(\mathbf{x})$ is the magnetic moment of unit volume. Relations (1.1)–(1.8) are given in the book of Landau and Lifshitz.^[1]

Magnetization of a magnet can be produced by another method; namely, by placing the magnet in the field of a superconducting ring or of a system of such rings. In this case, as is well known, the flux through the ring remains constant, whereas the superconducting current in the ring can change upon introduction of the magnet. We shall show that in this case the free energy is the quantity

$$\mathcal{F} = \mathcal{F} + \int [\mathbf{H}(\mathbf{x}) \mathbf{B}(\mathbf{x})/4\pi] d^3x \quad (1.9)^*$$

$$\delta \mathcal{F} = (4\pi)^{-1} \int \mathbf{H}(\mathbf{x}) \delta \mathbf{B}(\mathbf{x}) d^3x, \quad (1.10)$$

where the integration is carried out over the volume outside the superconductor (inside the superconductor, $\mathbf{B} = 0$).

By introducing the potential $\varphi(\mathbf{H} = -\nabla\varphi)$ and using the fact that $\text{div } \mathbf{B}(\mathbf{x}) = \text{div } \delta \mathbf{B}(\mathbf{x}) = 0$, we can put the integrand in (1.10) into the form

$$-\text{div}(\varphi \delta \mathbf{B}(\mathbf{x})) + \varphi \text{div } \delta \mathbf{B}(\mathbf{x}) = -\text{div}(\varphi \delta \mathbf{B}(\mathbf{x})).$$

The potential φ is a many-valued function; it changes by $4\pi I/c$ upon going around the current I along a closed contour that passes through the aperture of the ring. By using the fact that $\mathbf{B}_n = 0$ on the surface of the superconductor, it is easy to see that the integral in (1.10) reduces to an integral over the two sides of a surface capping the ring, and that it is equal to $I\delta Q/c$, where Q is the flux through the ring. In the general case,

$$\delta \mathcal{F} = c^{-1} \sum_i I_i \delta Q_i.$$

Thus the quantity \mathcal{F} actually is the thermodynamic potential in the variables Q_i , and for given fluxes Q_i it must be a minimum.

Besides this quantity, it is convenient to introduce also the total free energy \mathcal{F}_{tot} for given fluxes Q_i :

$$\mathcal{F}_{\text{tot}} = \mathcal{F} - \int [\mathbf{H}'_0(\mathbf{x})/8\pi] d^3x, \quad \delta \mathcal{F}_{\text{tot}} = (4\pi)^{-1} \int (\mathbf{H}\delta\mathbf{B} - \mathbf{H}'_0 \delta \mathbf{H}'_0) d^3x, \quad (1.11)$$

where $\mathbf{H}'_0(\mathbf{x})$ is the field produced by the system of

* $[\mathbf{H}\mathbf{B}] \equiv \mathbf{H} \times \mathbf{B}$.

superconducting rings, with the fluxes through them frozen, in a vacuum.

We can express the integrand in the second formula (1.11) in the form $\mathbf{H} \cdot \delta \mathbf{B} - \mathbf{H}'_0 \cdot \delta \mathbf{H}'_0 = (\mathbf{H} - \mathbf{B}) \cdot \delta \mathbf{H}'_0 + (\mathbf{B} - \mathbf{H}'_0) \cdot \delta \mathbf{H}'_0 + \mathbf{H} \cdot (\delta \mathbf{B} - \delta \mathbf{H}'_0)$. On setting $\delta \mathbf{H}'_0 = -\nabla \delta \varphi'_0$ and $\mathbf{H} = -\nabla \varphi$, we get

$$\begin{aligned} (\mathbf{B} - \mathbf{H}'_0) \delta \mathbf{H}'_0 &= -\text{div}[\delta \varphi'_0 (\mathbf{B} - \mathbf{H}'_0)], \\ \mathbf{H} (\delta \mathbf{B} - \delta \mathbf{H}'_0) &= -\text{div}[\varphi (\delta \mathbf{B} - \delta \mathbf{H}'_0)]. \end{aligned}$$

By using the fact that

$$\int (\mathbf{B} - \mathbf{H}'_0) d\mathbf{S} = \int (\delta \mathbf{B} - \delta \mathbf{H}'_0) d\mathbf{S} = 0,$$

where the integration extends over a surface covering the aperture of the superconducting ring, it is easy to see that the only contribution to $\delta \mathcal{F}_{\text{tot}}$ comes entirely from the term $(\mathbf{H} - \mathbf{B}) \cdot \delta \mathbf{H}'_0$; that is, that

$$\delta \mathcal{F}_{\text{tot}} = - \int \mathbf{M}(\mathbf{x}) \delta \mathbf{H}'_0(\mathbf{x}) dV. \quad (1.12)$$

An analogous relation holds also for the total free energy $\mathcal{F}'_{\text{tot}}$ of dielectrics:

$$\begin{aligned} \mathcal{F}'_{\text{tot}} &= \tilde{\mathcal{F}} + \int [\mathbf{E}'_0(\mathbf{x})/8\pi] d^3x, \\ \delta \mathcal{F}'_{\text{tot}} &= - \int \mathbf{P}(\mathbf{x}) \delta \mathbf{E}'_0(\mathbf{x}) dV. \end{aligned} \quad (1.13)$$

where $\mathbf{E}'_0(\mathbf{x})$ is the field produced in a vacuum at the given conductor potentials φ_i .

All the relations given above are valid for an arbitrary relation between \mathbf{H} and \mathbf{B} (or between \mathbf{E} and \mathbf{D}). In particular, this relation may be nonlocal (see Section c and also^[8]). If the relation between the field and the induction is local, then one can introduce a free-energy density as follows:

$$\begin{aligned} \tilde{F} &= -(4\pi)^{-1} \int_0^{\mathbf{E}} \mathbf{D} d\mathbf{E}, & F &= \tilde{F} + (4\pi)^{-1} \mathbf{E}\mathbf{D}, \\ \tilde{F} &= -(4\pi)^{-1} \int_0^{\mathbf{H}} \mathbf{B} d\mathbf{H}, & F &= \tilde{F} + (4\pi)^{-1} \mathbf{H}\mathbf{B}. \end{aligned}$$

The value of \tilde{F} is a minimum at given field (\mathbf{E} or \mathbf{H}), the value of F at given induction (\mathbf{D} or \mathbf{B}).

The following relations for the total free energies are also derived in the book^[1]:

$$\begin{aligned} \mathcal{F}_{\text{tot}} &= \int [F - (\mathbf{E}'_0/8\pi)] dV = \int [F - (\mathbf{E}\mathbf{D}/8\pi) - (1/2) \mathbf{P}\mathbf{E}_0] dV = \\ &= \int [\tilde{F} + (\mathbf{E}\mathbf{D}/8\pi) - (1/2) \mathbf{P}\mathbf{E}_0] dV, \quad (1.14) \end{aligned}$$

$$\begin{aligned} \tilde{\mathcal{F}}_{\text{tot}} &= \int [\tilde{F} + (\mathbf{H}'_0/8\pi)] dV = \int [F - (\mathbf{H}\mathbf{B}/8\pi) - (1/2) \mathbf{M}\mathbf{H}_0] dV = \\ &= \int [\tilde{F} + (\mathbf{H}\mathbf{B}/8\pi) - (1/2) \mathbf{M}\mathbf{H}_0] dV. \quad (1.15) \end{aligned}$$

In these formulas, the integrand vanishes outside the body, so that the integration extends only over the volume V of the body.

From formulas (1.3), (1.8), (1.12), and (1.13) it follows that the properties of a test particle (that is, a body which, because of its small dimensions, has practically no effect on the sources of the field) are independent of how the electrification (magnetization) is produced, provided the fields \mathbf{E}_0 and \mathbf{E}'_0 (or \mathbf{H}_0 and \mathbf{H}'_0) practically coincide inside the body. This result, however, was obvious beforehand.

b) Uniaxial and cubic ferromagnets. By way of illustration, we consider first the model of a uniaxial ferromagnet described in the book of Landau and Lifshitz.^[1] In this model, the free energy of unit volume of the ferromagnet is

$$\tilde{F} = F_0(\mathbf{M}) - (4\pi)^{-1} \int_0^H \mathbf{B} dH = F_0(\mathbf{M}) - \mathbf{M}\mathbf{H} - (H^2/8\pi). \quad (1.16)$$

The integral in formula (1.16) is calculated at a fixed value of the magnetic moment \mathbf{M} , which must then be found by minimization of \tilde{F} at given \mathbf{H} ; that is, from the equation $(\partial \tilde{F} / \partial \mathbf{M})_{\mathbf{H}} = 0$. The energy thus determined possesses the necessary property

$$\frac{\partial \tilde{F}}{\partial \mathbf{H}} = -\frac{\mathbf{B}}{4\pi} = -\frac{\mathbf{H}}{4\pi} - \mathbf{M}(\mathbf{H}),$$

where the differentiation of \tilde{F} is carried out with allowance for the $\mathbf{M}(\mathbf{H})$ dependence.

The quantity $F_0(\mathbf{M})$ is basically of exchange origin and in the first approximation is isotropic. Anisotropy appears only when relativistic interactions are taken into account:

$$F_0(\mathbf{M}) = F_0(|\mathbf{M}|) + U_{an}.$$

In the model under consideration, the magnetic anisotropy energy is

$$U_{an} = (1/2) \beta M^2 \sin^2 \theta, \quad \beta > 0,$$

where θ is the angle of inclination of the magnetic moment to the axis of easy magnetization (the z axis; $M_z = M \cos \theta$). In the plane perpendicular to this axis, there is no anisotropy in the present approximation. The relativistic origin of the anisotropy energy shows up in the fact that it is proportional to M^2 . The constant β in general is by no means small. In particular, it may be much larger than unity. The contrary case is also possible.

The absolute value of the magnetic moment \mathbf{M} may be considered constant. In this case $F_0(|\mathbf{M}|)$ is a constant, unimportant in the thermodynamics, and we shall hereafter omit it.

On minimizing \tilde{F} at given \mathbf{H} , we obtain an equation that determines the orientation of the magnetic moment:

$$\beta M \sin \theta \cos \theta = -H_z \sin \theta + H_x \cos \theta; \quad (1.17)$$

the magnetic field \mathbf{H} lies in the xz plane.

When $H_x^{2/3} + H_z^{2/3} < (\beta M)^{2/3}$, the free energy \tilde{F} as a function of the angle θ has two minima, of which one corresponds to absolute stability and the other to a metastable state. Thus in this case, two different values of \mathbf{M} (two phases) are possible for the same \mathbf{H} . In the region $H_x^{2/3} + H_z^{2/3} > (\beta M)^{2/3}$, metastable states are impossible, and consequently the direction of the magnetic moment \mathbf{M} at a given \mathbf{H} is uniquely determined.

The natural parameter for describing the amount of anisotropy is not β but

$$\beta/4\pi = [(\partial B_x / \partial H_x)_{H_z=0} - 1]^{-1}.$$

In the case of weak anisotropy this parameter is small in comparison with unity; and in the case of large anisotropy, large. The table below gives values of β , $\beta/4\pi$, M , and $\beta M^2/2$ for several uniaxial ferromagnets.

Substance	Parameter β	$\beta/4\pi$	M , gauss	$(\beta M^2)/2$, erg/cm ³
Co	4.2	0.33	1400	$4.1 \cdot 10^6$
MnSb	0.025	$2.10 \cdot 10^{-3}$	8900	$1 \cdot 10^6$
Mn ₂ Sb	0.06	$4.8 \cdot 10^{-3}$	2900	$0.25 \cdot 10^6$
Pfe ₁₋₂ O ₁₀ (magneto-plumbite)	43.4	3.45	330	$2.2 \cdot 10^6$

In cubic ferromagnets the anisotropy energy has the form

$$U_{an} = \beta' M^2 (m_1^2 m_2^2 + m_2^2 m_3^2 + m_3^2 m_1^2)/2,$$

where m_1, m_2, m_3 are the direction cosines of the vector \mathbf{M} with respect to three mutually perpendicular axes. When $\beta' > 0$, these axes are axes of easy magnetization. The constant β' is usually small in comparison with unity. In iron, $\beta' = 0.29$, $M = 1700$ G, $\beta' M^2/2 = 4.2 \times 10^5$ erg/cm³. Instead of equation (1.17) one obtains for cubic ferromagnets a more complicated equation, which we shall not write down. The maximum number of phases at a given field \mathbf{H} in cubic ferromagnets is six (in the case $\mathbf{H} = 0$).

In the nonuniform case there is added to the free energy a term dependent on the derivatives of the magnetization:

$$\tilde{\mathcal{F}} = \int \{ U_{an} - \mathbf{M}\mathbf{H} - (H^2/8\pi) + U_{nonunif} \} d^3x. \quad (1.18)$$

$$U_{nonunif} = (\alpha/2) (\partial M_i / \partial x_k)^2.$$

Because of the exchange origin of the nonuniformity energy, it is independent of the absolute direction of \mathbf{M} in the crystal. The constant α is usually of order 10^{-12} cm².

The relation between \mathbf{M} and \mathbf{H} in the nonuniform case is found from the condition that the free energy $\tilde{\mathcal{F}}$, considered as a functional of $\mathbf{M}(\mathbf{x})$ at fixed $\mathbf{H}(\mathbf{x})$, must be a minimum. One must allow for the fact that $|\mathbf{M}| = M = \text{const}$; that is, that the infinitely small variation $\delta \mathbf{M}(\mathbf{x})$ is perpendicular to $\mathbf{M}(\mathbf{x})$: $\delta \mathbf{M}(\mathbf{x}) = \delta \mathbf{a}(\mathbf{x}) \times \mathbf{M}(\mathbf{x})$. One can then write the condition for minimization of $\tilde{\mathcal{F}}$ in the form

$$\begin{aligned} [\mathbf{M}, \mathbf{H}_{\text{eff}}] &= 0, \\ \mathbf{H}_{\text{eff}} &= -(\delta \tilde{\mathcal{F}} / \delta \mathbf{M}(\mathbf{x}))_{\mathbf{H}(\mathbf{x})}. \end{aligned} \quad (1.19)$$

In uniaxial ferromagnets,

$$\mathbf{H}_{\text{eff}} = \mathbf{H} + \beta (M\mathbf{l})\mathbf{l} + \alpha \Delta \mathbf{M}, \quad (1.20)$$

where \mathbf{l} is the unit vector along the axis of easy magnetization (the z axis). This result was derived in^[13]. At the boundary between the ferromagnet and vacuum, the derivative of \mathbf{M} along the normal to the surface must vanish:

$$\partial \mathbf{M} / \partial n = 0.$$

This is also the condition for vanishing of the surface part of the variation of the free energy $\tilde{\mathcal{F}}$ (the natural boundary condition).

In view of the fact that the free energy $\tilde{\mathcal{F}}$ remains unchanged in infinitely small changes $\delta \mathbf{M}(\mathbf{x})$ that satisfy the condition $|\mathbf{M}| = \text{const}$ (that is, $\delta \mathbf{M} \perp \mathbf{M}$), the variation of the free energy can be expressed in the form (1.6); and consequently, all the necessary thermodynamic relations are satisfied in the model under consideration.

c) Conditions for coexistence of phases. On a boundary of separation between phases in magnets, the electrodynamic boundary conditions

$$\mathbf{H}_t = \text{const}, \quad B_n = \text{const} \quad (1.21)$$

must be satisfied; \mathbf{H}_t is the tangential component of the magnetic field, B_n is the normal component of the magnetic induction. These follow from the equations of magnetostatics $\text{curl } \mathbf{H} = 0, \text{div } \mathbf{B} = 0$. In addition, on a phase-separation boundary the thermodynamic condition

for coexistence of phases must be satisfied. To derive this condition, we note that the continuity of \mathbf{H}_t and \mathbf{B}_n at the phase-separation boundary plays the same role in our case as does the equality of temperatures and pressures in a liquid-vapor system. A natural requirement for equilibrium is equality of the thermodynamic potentials in the variables \mathbf{H}_t and \mathbf{B}_n . Since we are neglecting magnetostriction, this thermodynamic potential is the free energy

$$F' = \tilde{F} + (4\pi)^{-1} H_n B_n. \quad (1.22)$$

For given \mathbf{H}_t and \mathbf{B}_n , this quantity attains a minimum at equilibrium.

Thus the condition for equilibrium of phases has the form

$$F'_1(\mathbf{H}_t, B_n) = F'_2(\mathbf{H}_t, B_n), \quad F' = -(4\pi)^{-1} \int_0^H \mathbf{B} d\mathbf{H} + (4\pi)^{-1} H_n B_n. \quad (1.23)$$

The equality $F'_1 = F'_2$ means that the separation boundary is at a position of neutral equilibrium with respect to a displacement in the direction perpendicular to it. For ferroelectric materials, the analogous set of conditions has the form

$$F'(E_t, D_n) = \text{const}, \quad (1.24)$$

$$E_t = \text{const}, \quad D_n = \text{const}, \quad (1.25)$$

$$F'(E_t, D_n) = \tilde{F} + (4\pi)^{-1} E_n D_n = -(4\pi)^{-1} \int_0^{E_t} \mathbf{D} d\mathbf{E} + (4\pi)^{-1} E_n D_n. \quad (1.26)$$

The number of boundary conditions is four; this coincides with the number of independent variables—three components of magnetic (electric) field and one function $z(x, y)$ determining the position of the separation boundary. Therefore the magnetostatic (electrostatic) problem of subdivision into domains can be stated correctly as follows: it is required to find a solution of Maxwell's equations with the boundary conditions (1.21) to (1.23) or (1.24) to (1.25).

We emphasize that the derivation given above for the condition for coexistence of phases was not related to the specific nature of the magnetic (ferroelectric) material and is valid for all cases of coexistence of magnetic (ferroelectric) phases. These results were obtained in^[14] (see also^[8]).

In the case of coexistence of superconducting and normal phases, the boundary condition becomes simplified, since in the superconducting phase $\mathbf{B} = 0$. The problem of the intermediate state differs somewhat from the other problems of the theory of domain structure, although this difference is not one of principle. In the superconducting phase it is convenient not to introduce the vector \mathbf{H} at all (see, for example,^[11]). With this way of describing things, boundary conditions are imposed only in the normal phase, in which $\mathbf{H} = \mathbf{B}$ (the magnetization of a normal metal may be neglected). On the boundary with the superconductor, $H_n = 0$ (electrodynamic condition) and $H = H_C = \text{const}$ (thermodynamic condition). The problem of the intermediate state was solved by Landau^[15,16].

An important special case is a phase equilibrium such that on the separation boundary, not only is \mathbf{H}_t continuous, but so also is H_n . We recall that the condition $H_n = \text{const}$ does not follow from Maxwell's equations.

In this case the relations (1.21) to (1.26) can be rewritten as follows:

$$\mathbf{H} = \text{const}, \quad B_n = \text{const}, \quad \tilde{F} = -(4\pi)^{-1} \int_0^H \mathbf{B} d\mathbf{H} = \text{const}, \quad (1.27)$$

$$\mathbf{E} = \text{const}, \quad D_n = \text{const}, \quad \tilde{F} = -(4\pi)^{-1} \int_0^E \mathbf{D} d\mathbf{E} = \text{const}. \quad (1.28)$$

The equality $\tilde{F} = \text{const}$ follows from the conditions $F' = \text{const}$ and $H_n(E_n) = \text{const}$. The role of these boundary conditions will be elucidated in Chapter 3.

For the model of a uniaxial ferromagnet that was described above, the relation (1.27) means that the magnetic field is perpendicular to the axis of easy magnetization ($H_z = 0$) and is less in absolute value than βM (see^[11]). In this case, two equally stable states are possible:

$$M_{x1} = M_{x2} = H_x/\beta, \quad M_{y1} = M_{y2} = H_y/\beta, \\ M_{z1} = -M_{z2} = M [1 - (H/\beta M)^2]^{1/2}.$$

The phase-separation boundary must here be parallel to the axis of easy magnetization (this follows from the equality $B_{n1} = B_{n2}$, which in this case means that $M_{n1} = M_{n2}$). The orientation of the separation boundary in the plane perpendicular to the easy axis may be arbitrary.

We shall now discuss in more detail the conditions for phase coexistence in a uniaxial ferromagnet when $H_{n1} \neq H_{n2}$. In this case the equalities (1.21) to (1.23) can not be interpreted as simply as before, and we shall consider only the most important limiting cases.

1) **Ferromagnet with large anisotropy** ($\beta = \infty$). In this case the anisotropy energy is zero, since the magnetization \mathbf{M} does not deviate from the easy axis, and

$$F' = -M_z H_z - (H^2/8\pi) + (H_n B_n/4\pi), \\ M_{z1} = -M_{z2} = M.$$

Let the separation boundary form an angle ψ with the z axis, and let the y axis be chosen in the plane of the boundary (Fig. 1). If we use the fact that

$$-(H^2/8\pi) + (H_n B_n/4\pi) \\ = -(H^2/8\pi) + (8\pi)^{-1} [B_n^2 - (4\pi M_n)^2],$$

it is easy to see that this quantity must be the same on both sides of the separation boundary. Therefore the condition $F'_1 = F'_2$ takes the form $M_{z1} H_{z1} = M_{z2} H_{z2}$, whence it follows that

$$H_{z1} + H_{z2} = 0 \quad (\beta = \infty). \quad (1.29)$$

Since $H_z = H_t(x, z) \cos \psi - (B_n - 4\pi M_n) \sin \psi$, where $H_t(x, z)$ is the projection of \mathbf{H}_t on the xz plane, this condition is equivalent to the following:

$$H_t(x, z) = B_n \operatorname{tg} \psi \quad (\beta = \infty). \quad (1.30)$$

Either of these two relations, (1.29) or (1.30), can be

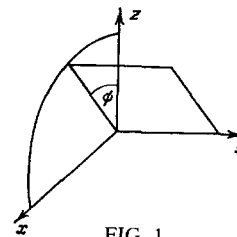


FIG. 1

used as the thermodynamic boundary condition. Thus if the separation boundary is inclined to the z axis, then on this boundary H_{z1} and H_{z2} do not vanish and $H_{x1} \neq H_{x2}$. The converse statement is also valid.

Formulas (1.29) and (1.30), which we obtained under the assumption that $\beta = \infty$, are actually valid also when the inequalities $\beta/4\pi \gg 1$ and $H \ll \beta M$ are satisfied.

The analogous formulas are correct for uniaxial ferroelectric materials in which the direction and magnitude of the polarization vector \mathbf{P} do not change even in a strong field \mathbf{E} . In such ferroelectric materials, the thermodynamic boundary condition has the form

$$E_{z1} + E_{z2} = 0,$$

or

$$E_{t(x,z)} = D_n \operatorname{tg} \psi.$$

2) Ferromagnets with small anisotropy ($\beta/4\pi \ll 1$).

In this case phase coexistence is possible in fields $H \lesssim \beta M \ll 4\pi M$. Therefore the condition $B_n = \text{const}$ can be written, in the zeroth approximation with respect to the parameter $\beta/4\pi$, in the form

$$M_n = \text{const.} \quad (1.31)$$

For simplicity, we shall consider only the case

$$H_{y1} = H_{y2} = 0, \quad M_{x1} = M_{x2} = 0$$

and shall suppose that the boundary plane is parallel to the y axis and forms an angle ψ with the axis of easy magnetization (Fig. 2). The angles formed by the vectors \mathbf{M}_1 and \mathbf{M}_2 with the z axis are equal respectively to θ_1 and to $-\theta_2$; it then follows from the condition (1.31) that $\psi - \theta_1 = \pi - \psi - \theta_2$; that is, $\theta_2 - \theta_1 = \pi - 2\psi$. The independent parameters may be considered to be, for example, the angles θ_1 and θ_2 . The four quantities H_{x1} , H_{x2} , H_{z1} , and H_{z2} are connected by four relations: two relations are obtained from the equation of state of the magnet (1.17) with $\theta = \theta_1$ and with $\theta = \theta_2$;* two other relations are the conditions $H_t = \text{const}$ and $F'(H_t, B_n) = \text{const}$. Thus for given \mathbf{M}_1 and \mathbf{M}_2 , the field at the phase-separation boundary is determined uniquely.

It must be noted, however, that the system of four equations mentioned above does not have, for all values of the parameters θ_1 and θ_2 , a solution corresponding to the coexistence of stable phases. In particular, the so-called 90-degree boundary ($\theta_1 = \pi/2$, $\theta_2 = \pi$, $\psi = \pi/4$) in a uniaxial ferromagnet with small anisotropy can not be realized.* In fact, on substituting in equation (1.17) the values $\theta = \theta_1$ and $\theta = -\theta_2$, we get the two relations

$$H_{z1} = H_{z2} = 0.$$

The condition $H_{t1} = H_{t2}$ in this case has the form

$$H_{x1} = H_{x2}.$$

The free energy F' can be expressed in the form

$$F' = U_{an} - \mathbf{M}_t \cdot \mathbf{H}_t - (H_t^2/8\pi) + (H_n^2/8\pi),$$

*Usually the relation (1.17) is considered an equation for \mathbf{M} at given \mathbf{H} . It is the condition for an extremum (with respect to \mathbf{M}) of the free energy $\tilde{F}(\mathbf{M}, \mathbf{H}) = U_{an} - \mathbf{M} \cdot \mathbf{H} - (H^2/8\pi)$. It must be remembered that the values of θ at which the free energy \tilde{F} has a maximum correspond to absolutely unstable states and must be rejected.

*On this point an error was committed in reference [14].

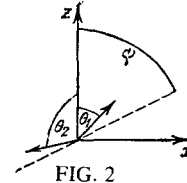


FIG. 2

the last two terms may be neglected, since $H \sim \beta M$. Since $\mathbf{M}_{t1} \cdot \mathbf{H}_{t1} = MH_{x1}/2$ and $\mathbf{M}_{t2} \cdot \mathbf{H}_{t2} = -MH_{z2}/2$, it is easy to see that from the condition $F'_1 = F'_2$ there follows

$$H_{x1} = H_{z2} = \beta M/2.$$

The state $\mathbf{M}_{x1} = M$, $H_{x1} = \beta M/2$, $H_{z1} = 0$ corresponds, as is easy to prove, not to a minimum but to a maximum of the free energy \tilde{F} at given \mathbf{H} ; that is, it is absolutely unstable. This means that in a uniaxial ferromagnet with small anisotropy, a 90-degree boundary is impossible.^[17]

In many textbooks on ferromagnetism (see also^[13]), a 90-degree boundary with $H_1 = H_2 = 0$ is considered. On such a boundary, the condition $F'_1 = F'_2$ is not satisfied; and one of the phases, the one whose magnetization is perpendicular to the easy axis, is absolutely unstable. Such a boundary, also, cannot be realized.

The range of values θ_1 and θ_2 in which the coexistence of stable phases is possible has dimensions (in the $\theta_1\theta_2$ plane) of order unity. The boundaries of this region have not been found in analytic form.

In cubic ferromagnets, a 90-degree boundary is possible; the magnetizations \mathbf{M}_1 and \mathbf{M}_2 in this case are directed along mutually perpendicular easy axes, so that $H_1 = H_2 = 0$ and $\tilde{F}_1 = \tilde{F}_2 = F'_1 = F'_2 = 0$. Such a boundary was considered in papers of Lifshitz^[18] and Néel^[19,20] and has been frequently observed in iron (see, for example, the review^[10]).

3) Angle between the boundary and the axis of easy magnetization small (β arbitrary). We shall give without derivation the thermodynamic boundary condition for this case, correct through terms of order ψ^2 . It has the form

$$H_{z1} + H_{z2} = 0 \quad (\psi \rightarrow 0),$$

the same as in the case $\beta = \infty$ (see (1.29)); but this time it is valid only in the second order with respect to ψ .

d) Structure of domain boundaries. Surface tension. We shall now consider the problem of the transitional layer between domains, assuming that the thickness of this layer is large in comparison with the distance between atoms (this situation occurs in all cases of coexistence of magnetic phases). It will be shown, in particular, that the problem of the transition layer has a solution only when the condition (1.23) for coexistence of phases is satisfied. This problem is one-dimensional; and in consequence of Maxwell's equations, the values of H_t and B_n do not change in the transition layer. The values of B_t and H_n can change in the direction perpendicular to the separation boundary (along the ξ axis). Far from the separation boundary (for $\xi \rightarrow \pm\infty$), they must approach the asymptotic values B_t^\pm and H_n^\pm . The orientation of the separation boundary with respect to the crystallographic axes we here consider arbitrary.

The free energy in the case under consideration is a functional of the distribution $B_t(\xi)$ and $H_n(\xi)$:

$$\tilde{\mathcal{F}} = \int_{-\infty}^{\infty} \tilde{F}\{B_t(\xi), H_n(\xi)\} d\xi.$$

A specific form of this functional is so far not necessary for our purposes.

We introduce also the free energies

$$\mathcal{F} = \tilde{\mathcal{F}} + (4\pi)^{-1} \int_{-\infty}^{\infty} H(\xi) B(\xi) d\xi$$

and

$$\mathcal{F}' = \tilde{\mathcal{F}} + (4\pi)^{-1} \int_{-\infty}^{\infty} H_n(\xi) B_n d\xi = \int_{-\infty}^{\infty} F'\{H_n(\xi), B_t(\xi)\} d\xi. \quad (1.32)$$

The values of B_n and H_t are connected with $\tilde{\mathcal{F}}$ and \mathcal{F} as follows:

$$B_n(\xi)/4\pi = -(\delta\tilde{\mathcal{F}}/\delta H_n(\xi))_{H_t(\xi)}, \\ H_t(\xi)/4\pi = (\delta\mathcal{F}/\delta B_t(\xi))_{B_n(\xi)}.$$

It is easily seen that these equations are the Euler-Lagrange equations for the functional \mathcal{F}' under the supplementary conditions $B_n = \text{const}$ and $H_t = \text{const}$. If there is a functional relation between $B_t(\xi)$ and $H_n(\xi)$, then one of these equations is a consequence of the other two.

In order that the functional \mathcal{F}' may have an extremum, it is necessary that the integrand in (1.32) take a unique value at $\xi \rightarrow \pm\infty$. Thus we again obtain the condition for coexistence of phases $F'_1(H_t, B_n) = F'_2(H_t, B_n)$.

This investigation shows that the structure of the transition layer can be found^[14] by minimization of the free energy \mathcal{F}' .

The analogous statement of the problem for ferroelectric materials has meaning in the vicinity of the Curie point. Far from the Curie point, the change of D_t and of E_n at the phase-separation boundary occurs in distances of the order of interatomic distances; that is, the problem of the structure of a domain wall loses meaning.

The surface tension Δ is the contribution of the domain wall to the free energy $\tilde{\mathcal{F}}$ (here we suppose that the currents $j(\mathbf{x})$ that produce the magnetic field are given). If on the phase-separation boundaries, besides the conditions (1.21) and (1.23), the condition $H_{n1} = H_{n2}$ is also satisfied (inside the wall, H_n may vary as usual), then from the equality $F'_1 = F'_2$ it follows that $\tilde{F}_1 = \tilde{F}_2$ (see (1.27)). In this case the surface tension can be defined as follows:

$$\Delta = \int_{-\infty}^{\infty} [\tilde{F}\{B_t(\xi), H_n(\xi)\} d\xi - \tilde{F}(\xi = \pm\infty)].$$

But if $H_{n1} \neq H_{n2}$, then also $\tilde{F}_1 \neq \tilde{F}_2$, and the value of Δ cannot be defined in such a manner. In fact, the position of the separation boundary is defined only to within the wall thickness δ . Therefore in the case $\tilde{F}_1 \neq \tilde{F}_2$ it is impossible uniquely to isolate from the quantity $\int \tilde{F}\{B_t(\xi), H_n(\xi)\} d\xi$ that part that is due to the formation of a domain boundary. The indeterminacy in the surface energy is of the order of $\delta(\tilde{F}_1 - \tilde{F}_2)$. If this quantity is small, then the surface tension can be defined approximately. Otherwise, the energy due to formation of a domain boundary becomes nonlocal, and the concept of surface tension loses meaning.^[17]

In uniaxial ferromagnets, the condition $\tilde{F}_1 = \tilde{F}_2$ is satisfied only for boundaries parallel to the axis of easy magnetization. For large angles of deviation of the

boundary, the surface tension cannot be defined by any reasonable method.

In superconductors, this difficulty is absent, since on the separation surfaces $B_n = 0$ and consequently $\tilde{F}_1 = \tilde{F}_2$.

The simplest and at the same time most important case of phase coexistence in uniaxial ferromagnets was investigated in the papers of Bloch^[21] and of Landau and Lifshitz^[13]. In this case, the magnetizations of the coexisting phases are opposite in direction and parallel to the easy axis, and the field \mathbf{H} is zero. Inside the transition layer, the magnetization \mathbf{M} rotates about the normal to the separation boundary (the x axis), remaining parallel to the plane of the boundary ($M_z = M \cos \theta$, $M_y = M \sin \theta$, $M_x = 0$). The function $\theta(x)$ is determined by minimization of the functional (see (1.18))

$$\tilde{\mathcal{F}} - \mathcal{F} = \int_{-\infty}^{\infty} \left[\frac{\beta}{2} M^2 \sin^2 \theta + \frac{\alpha}{2} (\partial M / \partial x)^2 \right] dx \\ = \int_{-\infty}^{\infty} \left[\frac{\beta}{2} M^2 \sin^2 \theta + \frac{\alpha}{2} M^2 (\partial \theta / \partial x)^2 \right] dx \quad (1.33)$$

under the conditions $\theta(-\infty) = 0$, $\theta(+\infty) = \pi$. The Euler-Lagrange equation for this problem has the form

$$\alpha \theta'' - \beta \sin \theta \cos \theta = 0.$$

After simple calculations, one obtains

$$\cos \theta = -\text{th}(x/\delta),$$

where $\delta = (\alpha/\beta)^{1/2}$ is the thickness of the domain boundary.

The surface tension Δ is equal to the integral (1.33):

$$\Delta = 2(\alpha\beta)^{1/2} M^2 = 2\beta\delta M^2. \quad (1.34)$$

The domain wall in crystals of cubic symmetry was investigated in^[18,19]. The surface tension of a 180-degree boundary parallel to a plane of type (001) in a cubic crystal is

$$\Delta_{180^\circ} = (\alpha\beta')^{1/2} M^2.$$

The thickness of a 180-degree boundary in this case is determined by magnetostriction and is appreciably larger than $(\alpha/\beta')^{1/2}$. In iron, $\Delta_{180^\circ} = 1.8 \text{ erg/cm}^2$.^[10]

In cubic ferromagnets, 90° boundaries are often observed: $\mathbf{H} = 0$, the magnetizations \mathbf{M}_1 and \mathbf{M}_2 are parallel to the crystallographic axes [100] and [010], and the plane of the boundary is the (110) plane. The energy of such a boundary is^[19]

$$\Delta_{90^\circ} = 0.863 (\alpha\beta')^{1/2} M^2,$$

and the thickness of the transition region is of order $(\alpha/\beta')^{1/2}$.

The structure of a domain boundary in ferroelectric materials near the Curie point was described in the review.^[22] The transition region in superconductors was investigated by Ginzburg and Landau.^[23] Other types of domain boundaries were considered in^[8].

Certain specific properties are possessed by phase-separation boundaries in thin metal films, whose thickness is comparable with the thickness of the transition layer. We shall not consider this case. The theory of thin magnetic films is set forth, for example, in the books^[24].

2. DOMAIN STRUCTURES IN FERROMAGNETS

a) **Simplest domain structures.** We shall first explain why a division into domains occurs in massive specimens. We shall consider as an example a plane-parallel ferromagnetic plate of thickness l , cut perpendicularly to the easy axis (the ferromagnet is assumed to be uniaxial). We shall suppose that the dimensions of the plate in the plane perpendicular to the easy axis are infinite and that the anisotropy is extremely large ($\beta/4\pi \rightarrow \infty$). In addition, we shall suppose for simplicity that there is no external field ($H_0 = 0$). In such a plate, a uniform state is possible: magnetization M parallel to the easy axis, internal field $H = -4\pi M$. The free energy per unit volume in this case is $F^{unif} = \tilde{F}^{unif} = 2\pi M^2$. The state considered is metastable, since H and M are antiparallel (in the case of small anisotropy, this state would be absolutely unstable), and it is obvious there exists a structure with smaller free energy. Such a structure is the so-called Kittel structure^[25] shown in Fig. 3. The arrows show the directions of the magnetization. The free energy of this structure consists of two parts: the energy of surface tension on the phase-separation boundaries (this energy, per unit area of the plate, is $\Delta l/a = 2\beta\delta M^2 l/a$) and the energy of emergence of the domains to the surface. The latter is due to the fact that near the specimen surface (at distances of the order of the domain width a), there is a nonuniform magnetic field $H \sim 4\pi M$ (in general, the emergence energy includes also anisotropy energy). By symmetry it is evident that the field H on the phase-separation boundaries is perpendicular to the separation boundaries, so that not only the conditions (1.21) and (1.23) for coexistence of phases but also the more exacting equations (1.27) are satisfied. The magnetic field distribution is calculated, for example, in the book of Landau and Lifshitz.^[1] The energy of emergence of the domains to the specimen surface, per unit area of the plate (with allowance for the two sides of the plate), is $1.7 M^2 a$. The domain width a is determined by minimization of the sum

$$2\beta\delta M^2 (l/a) + 1.7 M^2 a,$$

that is,

$$a \approx 1.1 (\beta\delta l)^{1/2}.$$

The total free energy of the domain structure shown in Fig. 3 increases, on increase of the plate thickness l , in proportion to $l^{1/2}$, whereas the free energy of the uniform state is proportional to the first power of l . It is therefore clear that a division into domains will be energetically advantageous at sufficiently large specimen dimensions ($l > l_c \sim \beta\delta$).

The role of the energy of surface tension and of the energy of emergence of the domains to the surface was demonstrated (for another model) in the paper of Landau and Lifshitz.^[13] In the same paper, the dependence of

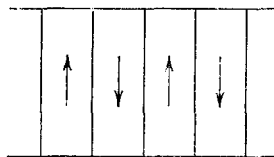


FIG. 3

the domain width a on the plate thickness l was derived; it has the form $a \sim l^{1/2}$.

In an external magnetic field H_0 , perpendicular to the specimen surface, the concentrations of the phases change, and the layer boundaries must bend near the surface,^[14] just as in superconductors.^[1] If the boundaries were to remain straight, the conditions for phase coexistence, $F'(H_t, B_n) = \text{const}$, would not be satisfied (for a given distribution of magnetization $M(x)$, the magnetostatic problem has a unique solution, and it is not possible to impose an additional condition). The curvature of the boundaries near the specimen surface is of the order of $H_0/4\pi M$ (the magnetostatic problem contains no other dimensionless parameters, since $\beta/4\pi = \infty$).

The simple structure shown in Fig. 3 is actually observed only in quite thick plates (see, for example,^[26,27]); and for large dimensions l , a complication of this structure occurs (see the next section), and the dependence of a on l changes. But the basis of the structures that do form is, as before, a division into two phases with opposite magnetizations.

In cubic crystals with a positive anisotropy constant β' , in zero external field, there can occur a structure with closed flux, suggested by Lifshitz^[18] (Fig. 4). The emergence energy in this case is magnetostrictive energy, which is proportional to the volume of the closure prisms; that is, its amount per unit area of the plate surface is proportional to the domain width a . The magnetostriction energy can be estimated^[18] by ascribing to the triangular domains an effective uniaxial anisotropy energy $U = kM^2$. In iron, $k = 3.3 \times 10^{-4}$. Thus is obtained an upper bound for the magnetostrictive energy of the whole body.^[18] The energy of the structure shown in Fig. 4, per unit area of the plate surface, is

$$E = (kM^2 a/2) + (\Delta_{180} l/a).$$

On minimizing this expression, we obtain the dependence of a on l :

$$a = (2\Delta_{180} l/kM^2)^{1/2}.$$

In the case of iron

$$a = 6.2 \cdot 10^{-2} l^{1/2} \text{ cm}$$

For the ratio $a/l^{1/2}$ we obtain an estimate that bounds it from below, since the magnetostriction energy is actually less than $kM^2 a/2$.

Strictly speaking, in an infinite plate such a structure is metastable, and a preferable state would be a uniform one with magnetization oriented along an easy axis that is parallel to the plane of the plate. But in even a small external field perpendicular to the plane of the plate, the structure shown in Fig. 4 becomes energetically advantageous. In the absence of a field, this structure can be energetically advantageous in a plate of finite dimensions, if the ends of the plate are parallel to an

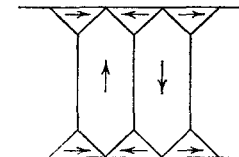


FIG. 4

easy axis. The structure shown in Fig. 4 has often been observed in iron (see, for example, [10,12,28]). A curious structure is that of Néel (Fig. 5) [20], which is observed in iron strips cut so that the length of the strip coincides with a [110] direction. The method of magnetic flux closure shown in Fig. 5 was suggested by Lawton (thesis, Cambridge, 1949) and was described in [28].

In many textbooks it is supposed that the structure with closed flux (internal field $H = 0$) shown in Fig. 4 is also realized in uniaxial ferromagnets with small anisotropy. Actually such a structure is impossible, even if it is supposed that the magnetization in the triangular closure domains is turned from the easy axis by an internal field $H \sim \beta M$. [17] In fact, a 90-degree boundary cannot be realized: the phase-coexistence condition $F'(\mathbf{H}_t, \mathbf{B}_n) = \text{const}$ can be satisfied only if one of the phases is absolutely unstable (see Section c of Chapter 1).*

The conditions for coexistence of phases can be obtained from the equations of the microtheory (see Section d of Chapter 1). In the present model these are the static equations (1.19) and (1.20) of Landau and Lifshitz, which are themselves derived from the condition that the free energy \mathcal{F} be a minimum. Therefore a structure that does not satisfy the boundary condition $F'_1 = F'_2$ does not correspond to an energy minimum.

The problem of the domain structure of uniaxial ferromagnets with small anisotropy has still not been solved. The paper [17] presents a rather complicated model of such a structure and discusses the difficulties of the theory (see also the following section). It is obvious, however, that in the zeroth approximation with respect to the parameter $\beta/4\pi \ll 1$, the flux must close within the specimen; that is, on the specimen surface the component M_n of the magnetization normal to the surface must vanish. Since the magnetization \mathbf{M} can deviate from the easy axis only in the presence of an internal field \mathbf{H} , it is obvious that near the surface of

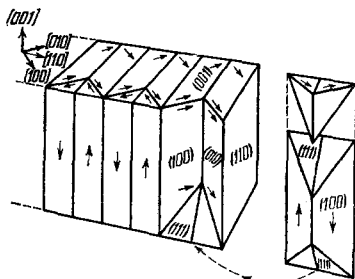


FIG. 5

*In the literature it is often assumed that formation of locally unstable states is necessary for attainment of a minimum of the free energy of the whole body, \mathcal{F} . The fact is cited that Maxwell's equations make the problem nonlocal. But it can be shown that the condition for a minimum of the free energy \mathcal{F} leads to a local condition that the free-energy density \bar{F} , considered as a function of \mathbf{M} at given local value of \mathbf{H} , be a minimum. In order to prove the instability of the triangular closure domains, it is sufficient to consider only infinitely small perturbations of the form $\delta\mathbf{H} = 0$, $\delta\mathbf{M} = (4\pi)^{-1} \text{curl } \delta\mathbf{A}$, $\delta\mathbf{A} = \{\delta A(\mathbf{x}), 0, 0\}$, where $\delta A(\mathbf{x})$ is an arbitrary function of its three variables ($\delta M_z \neq 0$), localized within the triangular domains, and the x axis is chosen along the direction of \mathbf{M} in the triangles. Such a perturbation decreases the anisotropy energy without disturbing the equations $\text{curl } \mathbf{H} = 0$ and $\text{div } \mathbf{B} = 0$.

the specimen there is a nonuniform field $H \sim \beta M$ [14,17], which must orient the magnetization almost perpendicular to the easy axis. The component of this field perpendicular to the easy axis and consequently parallel to the surface must differ from zero. This field will penetrate beyond the boundaries of the specimen (to distances of the order of the domain width a) and can be measured. We remark that fields of the order of 10^4 Oe have been frequently observed over the surface of monocrystals of cobalt, [29] in which $\beta/4\pi \approx 1/3$, and cannot be explained within the framework of the generally accepted theory, in which deviation of the magnetization from the easy axis is allowed in the absence of an internal field H .

c) Plane model of the branching of the domain structure of uniaxial ferromagnets. With increase of the plate thickness l , a progressive branching of the domains in the vicinity of the surface becomes thermodynamically advantageous. This was first observed by Landau in an investigation of the intermediate state of superconductors. [16] The initial stage of branching in ferromagnets was considered in a paper of Lifshitz. [18]

In uniaxial ferromagnets, in contrast to superconductors, branching of the domains becomes advantageous at rather small dimensions l . Therefore great interest attaches to the problem of the extremely branched structure. A plane model of such a structure was constructed in [17] for the case of a uniaxial ferromagnet with small anisotropy. It was shown that the dependence of the layer width a on the plate thickness l changes: $a \sim l^{2/3}$. This dependence was observed in cobalt (see, for example, [26,30]). The same dependence was obtained by Landau for superconductors. [18]

We shall formulate the magnetostatic problem of the emergence of domains to the surface. We suppose that there is no external magnetic field and that the axis of easy magnetization is perpendicular to the plane of the ferromagnetic plate. The energy density of emergence is

$$F = U_{\text{an}} + (H^2/8\pi) = (1/2) \beta M^2 \sin^2 \theta + (H^2/8\pi).$$

In the limit of small anisotropy, the second term may be neglected, since $H \sim \beta M \sin \theta$. Thus in the first approximation, the field \mathbf{H} may be neglected, and the equations of magnetostatics reduce to the single equation

$$\text{div } \mathbf{M} = 0 \quad (|\mathbf{M}| = M = \text{const}).$$

From the mathematical point of view, the problem reduces to solution of the eikonal equations

$$\left(\frac{\partial A}{\partial x}\right)^2 + \left(\frac{\partial A}{\partial z}\right)^2 = 1, \quad \frac{\mathbf{M}}{M} = \text{rot } \mathbf{A}, \\ \mathbf{A} = \{0, A(x, z), 0\}.$$

The lines of force are the lines of equal values of the vector potential \mathbf{A} .

A model of the branched structure is shown in Fig. 6. At a depth h , which will be calculated below, each domain splits. With approach to the surface, the width of the new domains increases, until it becomes equal to $a/3$. At this point a new splitting occurs. The process continues until the dimensions of the domains that are forming becomes comparable with the thickness δ of a domain wall.

The concentration of the opposite phase in an orig-

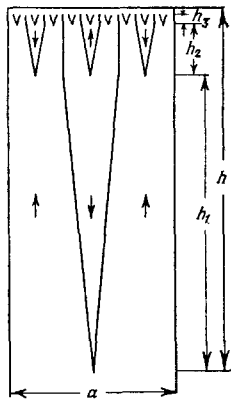


FIG. 6

inal domain (of width a) after the n -th splitting is determined by the recurrence relation

$$c_n = (2/3)c_{n-1} + (1/3)(1 - c_{n-1}); \quad c_1 = 1/3,$$

that is

$$c_n = (1/3) + (1/9) + \dots + (1/3^n).$$

For $n \rightarrow \infty$ we get $c_\infty = 1/2$. This means closure of the magnetic flux within the specimen; that is, vanishing of the component of the magnetization normal to the surface (averaged over distances of the order of δ).

A schematic drawing of the splitting is shown in Fig. 7. The fine solid lines represent the lines of force. The heavy lines show the phase-separation boundaries. The lines of force consist of sections of straight lines and of arcs of circles whose centers are at the points O , O' , and O'' . In the central domain, the lines of force are parallel to the axis of easy magnetization. The magnetization M is parallel (or antiparallel) to the easy axis at the beginning and at the end of each stage of splitting.

From the flux-conservation condition it follows that a point on the separation boundary must be equally distant from the straight line of force emanating from the point O' , and from the arc passing through the center of the figure, which is the continuation of that straight line in its role as a line of force (in the latter case, what is meant is the distance along a radial line). Therefore

$$a_n/3 = R_n [(\cos \theta_n)^{-1} - 1].$$

We have in addition

$$h_n = R_n \operatorname{tg} \theta_n.$$

It will be shown below that the angles θ_n are small (of the order of $(\delta/a_n)^{1/2}$). Therefore

$$\begin{aligned} R_n &= 2a_n/3\theta_n^2, \\ h_n &= 2a_n/3\theta_n, \end{aligned} \quad (2.1)$$

in which

$$a_n = a/3^{n-1},$$

The parameters R_n , h_n , and θ_n are determined by the minimization condition for the total energy connected with the splitting. The anisotropy energy in the section of height h_n is

$$U^{(n)} = \beta M^2 \int_0^{\theta_n} \sin^2 \theta d\theta \left[\int_{[R_n - (a_n/2)] \cos \theta}^{R_n / \cos^2(\theta/2)} r dr + \int_{a_n/6 \cos^2(\theta/2)}^{a_n/2 \cos \theta} r' dr' \right].$$

By using (2.1) and also the fact that $\theta_n \ll 1$, and by neglecting terms of order $a_n^2 \theta_n^3$, we get

$$U^{(n)} = (4/45) \beta M^2 a_n^3 \theta_n.$$

The surface-tension energy $E^{(n)}$ is*

$$E^{(n)} = 3h_n \Delta,$$

where $\Delta = 2\beta\delta M^2$; δ is the thickness of the domain wall, i.e.,

$$E^{(n)} = 4\beta\delta M^2 a_n / \theta_n.$$

The angle θ_n is determined by the minimization condition for the sum $U^{(n)} + (2E^{(n)}/3)$. The second term includes only the energy of the boundaries of the central domain. Thus in a given step we shall minimize the energy that is connected with the splitting. After simple calculations we get

$$\theta_n^2 = 30\delta/a_n.$$

The total energy of the region of thickness a_n and height h_n is

$$F^{(n)} = U^{(n)} + E^{(n)} = (2\sqrt{10}/3\sqrt{3}) \beta M^2 a_n^{3/2} \delta^{1/2}.$$

By using

$$\sum_n a_n^{3/2} = a^{3/2} [1 - (1/3\sqrt{3})]^{-1}, \quad \sum_n 3^{n-1} a_n^{3/2} = a^{3/2} [1 - (1/\sqrt{3})]^{-1},$$

we get

$$\begin{aligned} \sum_n 3^{n-1} F^{(n)} &= (5/2) \lambda_1 \beta M^2 a^{3/2} \delta^{1/2}, \quad \lambda_1 = 8/3 \sqrt{10} (\sqrt{3} - 1) = 1.15; \\ h &= \sum_n h_n = 1/4 \lambda_2 (a^{3/2} \delta^{1/2}), \quad \lambda_2 = 8/\sqrt{10} (3\sqrt{3} - 1) = 0.60. \end{aligned}$$

The total energy of the specimen per domain of thickness a is

$$\begin{aligned} E &= 2 \sum_n 3^{n-1} F^{(n)} + 2\beta\delta M^2 (l - 2h) = 2\beta M^2 a (2\lambda_1 a^{1/2} \delta^{1/2} + \delta l a^{-1}), \\ \lambda &= (5\lambda_1 - \lambda_2)/4 = 1.29, \end{aligned}$$

where l is the plate thickness.

It is still necessary to minimize the energy per unit area of the plate surface, that is E/a . This gives

$$\begin{aligned} a &= \delta^{1/3} l^{2/3} / \lambda^{2/3}, \quad E/a = 6\lambda^{2/3} \beta M^2 \delta^{2/3} l^{1/3}, \\ h &= (\lambda_2/4\lambda) l = 0.117l, \quad h_1 = h [1 - (1/3\sqrt{3})], \quad h_n = h_l / (3\sqrt{3})^{n-1}, \\ \theta_n &= 3^{n/2} \sqrt{10} (\delta/\lambda l)^{1/3}. \end{aligned} \quad (2.2)$$

It is easily shown that

$$h - \sum_{n=1}^{n'} h_n = h / (3\sqrt{3})^{n'}.$$

If this quantity is of the order of δ , that is $(3\sqrt{3})^{n'} \sim l/8\delta$, then $a_n \sim \delta$, whereas $\theta_n \sim 1$. Thus the total number of splittings is

$$n_{\max} \sim (2/3) (\ln 3)^{-1} \ln (l/\delta).$$

Figure 6 corresponds to the case $l/\delta \sim 10^4$. It is seen that the number of splittings is still quite small. With increase of l , the ratio a/l decreases, that is the angle of inclination of the boundaries decreases, while the number of splittings increases.

*In Section d of Chapter I it was shown that, strictly speaking, the concept of surface tension loses meaning for boundaries inclined to the axis of easy magnetization, since the surface energy is defined only to within $\delta(\vec{F}_1 - \vec{F}_2)$. For boundaries parallel to the easy axis, $\vec{F}_1 = \vec{F}_2$. In the case under consideration, the inclination of the boundaries is small (of order θ_n), and the indeterminacy in the surface energy is small in comparison with Δ .

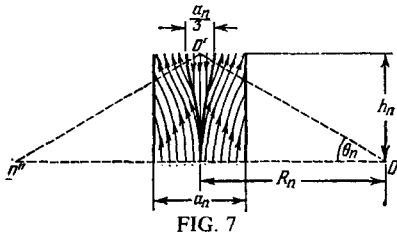


FIG. 7

The structure actually observed is not plane. On a surface perpendicular to the easy axis in a cobalt monocrystal, very complicated patterns are observed (see, for example, [28]). Similar patterns have been observed in Mn_2Sb . [31] Nevertheless the relation $a \sim l^{2/3}$ is satisfied. [26,30]

It is interesting to compare the energy of the branched structure (2.2) with the energy of the unbranched structure, which is obviously of the order of $\beta M^2 (\delta l)^{1/2}$. It is easily seen that the unbranched structure ceases to be stable for $l > l_c \sim C\delta$, where C is a numerical coefficient independent of β . In order to estimate this coefficient, it is necessary to know the energy of the unbranched structure accurately, since all the numerical factors occur to the sixth power in the ratio l_c/δ .*

In superconductors, according to estimates of Lifshitz and Sharvin, [32] the numerical coefficient $l_c/\delta \sim (50)^6$, and therefore the branched structure is never observed. In ferromagnets with small anisotropy, the situation is different. It follows from experiment that the multiplier C is not very large; in every case it is many orders smaller than in superconductors. Because the multiplier C does not contain literal parameters, the problem of the unbranched structure cannot be formulated correctly within the framework of macrotheory. [17] In particular, an important contribution to the energy of the unbranched structure is made by boundaries inclined to the axis of easy magnetization, and this contribution cannot be taken into account within the framework of macrotheory. The difficulties are due to the fact that the ratio l/a is not very large and does not contain large parameters of the type $4\pi/\beta$. To explain the properties of the unbranched structure would, strictly speaking, be possible only by solving the static Landau-Lifshitz equation (1.19) in the whole volume of the specimen, which seems to us impossible. As has already been indicated, the situation in superconductors is much more favorable. Although the ratio δ/l_c still contains no small parameters, it is nevertheless so small that the approximation being used occasions no doubts.

The results given above from investigation of the branched structure are valid also for ferromagnets of cubic symmetry. In this case the anisotropy energy for small deviations from the easy axis can be written in the form $U_{an} = (\beta' M^2 \theta^2)/2$. The surface-tension energy Δ can be expressed in the form $\Delta = 2\beta'\delta M^2$, where $\delta = [(\alpha/\beta)^{1/2}]/2$ is a coefficient of proportionality, not identical with the thickness of a domain wall. In cubic ferromagnets the critical dimension l_c is appreciably larger because of the fact that the energy of the un-

*Strictly speaking, in order to calculate l_c exactly it would be necessary to consider the initial stage of branching; that is, to investigate the stability of the unbranched structure with respect to infinitely small perturbations.

branched structure is very small. [18] By considering the initial stage of branching, Lifshitz [18] obtained for l_c the value

$$l_c = 8 (\beta'/k)^3 (\alpha/\beta')^{1/2}.$$

In iron this quantity is of order 10^4 cm. It is for this reason that cubic ferromagnets are especially suitable for observation of a simple domain structure with closure triangles.

It is also possible to treat the problem of branched domain structure in ferromagnets with an arbitrary anisotropy constant. In order to estimate the parameters of such a structure, we shall suppose that in this case also the picture shown in Fig. 6 is realized; that is, $a_{n+1}/a_n = 1/3$. This supposition is not in harmony with the thermodynamic boundary condition $H_{z1} + H_{z2} = 0$ (see Section c of Chapter 1), so that the method presented below allows us to find only an upper bound to the energy of the branched structure (as has already been mentioned, the thermodynamic boundary condition follows from the minimization condition for the free energy of the body, \mathcal{F}).

We suppose that at each stage of the splitting the boundaries of the central domain are parabolas $r = R_n/\cos^2(\theta/2)$ and $r' = a_n/6 \cos^2(\theta/2)$. The discontinuity of the normal component of magnetization on the phase-separation boundaries leads to the presence of a magnetic field in the side domains:

$$H_x(x, z) \approx 8\pi M \mu^{-1} \theta_0(z), \quad H_z \leq H_x, \quad \mu = 1 + 4\pi\beta^{-1}.$$

In the central domain, in a first approximation, $H_x = 0$. The emergence energy is the energy of the magnetic field, $\int (H^2/8\pi) dV$. We omit the calculation, which is analogous to that made in the case $\beta/4\pi \leq 1$, and give only the final result:

$$E/a = 6M^2 (\lambda\beta\delta)^{2/3} (4\pi l/\mu)^{1/3}, \quad a = (\beta\mu\delta/4\pi)^{1/3} (l/\lambda)^{2/3}, \quad h = 0.117l.$$

The relation $a \sim l^{2/3}$ has been observed also in magnetoplumbite. [26,27] By equating the energies of the branched and unbranched structures in the case $\beta/4\pi \gg 1$, it is possible to estimate the critical dimension l_c above which the branching is certainly advantageous: $l_c \approx 0.87 \times 10^4 \beta\delta$. The ratio a_c/l_c (where a_c is the domain dimension in the unbranched structure at $l = l_c$) is 1.2×10^{-2} . We note that the value obtained for l_c is too large and for the ratio a_c/l_c too small. In the initial stage of complication of the simple unbranched structure, there occurs a bending of the plane domain boundaries, in which the structure ceases to be two-dimensional, and thereafter nuclei of reversed magnetization appear near the surface. [26]

c) **Plane model of branched domain structure in cubic ferromagnets.*** The simplest generalization of a domain structure with closure triangles for a case in which the plate surface is inclined to the easy axis is the structure shown in Fig. 8, where

$$\delta_1 = (\pi/4) - (\gamma/2), \\ \delta_2 = (\pi/4) + (\gamma/2).$$

The anisotropy-energy density in the triangular domains is $U_{an} = (1/2)\beta' M^2 \sin^2 \gamma \cos^2 \gamma$. In iron, $\beta' = 0.28$. This structure is unbranched, so that $a \sim l^{1/2}$.

*The results described in this section were obtained in [33].

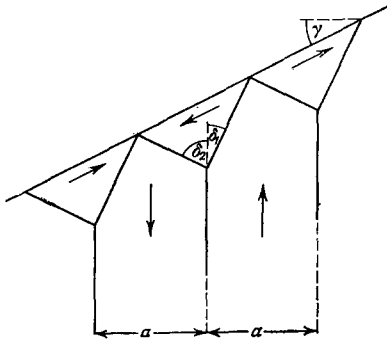


FIG. 8

In the case $\gamma \sim 1$, the structure shown in Fig. 8 is energetically disadvantageous. The total energy can be appreciably decreased by formation of the branched structure shown in Fig. 9. This figure shows only two successive stages of the branching. Actually the subdivision proceeds to the point at which the domain dimensions become comparable with the thickness δ_{180° of a 180-degree boundary (this is much larger than the thickness δ_{90° of a 90-degree boundary). This enables us to estimate the number of branchings:

$$n \sim \ln(a/\delta_{180^\circ})/\ln 3.$$

In this way we can get rid almost completely of the anisotropy energy. It is concentrated entirely in a thin layer close to the surface of the specimen (at distances of order δ_{180°). The energy of such a structure, per unit area of the plate surface (with allowance for the two sides of the plate), is

$$E = [(4/3 \ln 3) (2\sqrt{2} \Delta_{90^\circ} + \Delta_{180^\circ}) \ln(a/\delta_{180^\circ}) + (2kM^2 a/3) + \Delta_{180^\circ} (l - 2a) a^{-1}] \cos \gamma. \tag{2.3}$$

The term proportional to $\ln(a/\delta_{180^\circ})$ is the energy of the branched boundaries (Δ_{90° and Δ_{180° are the energies of 90-degree and 180-degree boundaries: $\Delta_{90^\circ} = 0.863 \times \Delta_{180^\circ} \sim \beta' \delta_{90^\circ} M^2$). After each branching the domain dimensions are halved, but their number increases correspondingly, and therefore the total energy of the branched boundaries is proportional to the number of branchings n . The branching is energetically advantageous if this energy is less than the anisotropy energy of the triangular domains shown in Fig. 8. For $\gamma \sim 1$ and $a \gtrsim \delta_{180^\circ}$, this condition is always satisfied.

The second term represents the magnetostriction

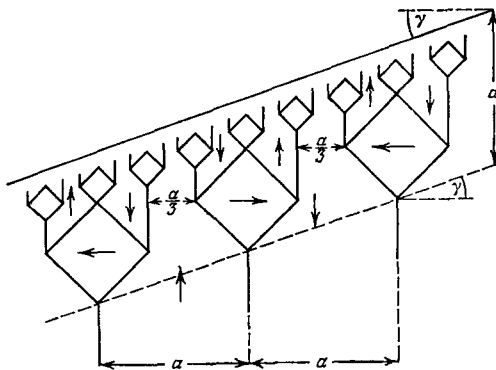


FIG. 9

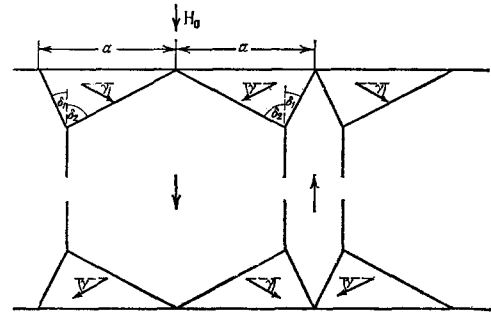


FIG. 10

energy. We estimate it by ascribing to the quadrangular domains an effective uniaxial anisotropy energy $U_{ms} = kM^2$. In iron, $k = 3.3 \times 10^{-4}$. As has already been indicated, one obtains thus an upper bound to the magnetostrictive energy of the whole body.^[18] The last term represents the surface-tension energy of the unbranched boundaries (the plate thickness l is measured along an easy axis). On minimizing this expression, we obtain a quadratic equation for a . The positive root of this equation has the form

$$a = 0.466l [1 + (1 + l_{c1}^{-1})^{1/2}]^{-1}, \tag{2.4}$$

$$l_{c1} = (2\sqrt{2} \Delta_{90^\circ} + \Delta_{180^\circ})/kM^2 \ln 3 \sim \delta_{90^\circ} \beta'/k \sim \delta_{180^\circ} \beta'/k \ln(\beta'/k). \tag{2.5}$$

In the case of iron, $l_{c1} = 6 \cdot 10^{-3}$ cm.

Thus for small thicknesses, $l \ll l_{c1}$, the relation $a = 0.233 l$ should be satisfied, whereas for $l \gg l_{c1}$ this becomes a square-root dependence, $a = 0.466 (l_{c1}/l)^{1/2}$, despite the fact that the structure is branched. In the latter case the energy is $E = 0.62 kM^2 (l_{c1}/l)^{1/2}$. At very large values of l , there should be observed an extremely branched structure with a $\sim l^{2/3}$, of the same type as that considered in the preceding section. The energy of such a structure is of the order of $\beta' \delta_{90^\circ}^{2/3} M^2 l^{1/3}$. By equating energies, we obtain for the critical thickness the value $l_c \sim (\beta'/k)^3 \Delta_{90^\circ}$.*

In the case of iron this value is of the order of 10^3 cm, so that the extremely branched structure is practically unattainable.

So far we have assumed that there is no external field. In the presence of an external field H_0 perpendicular to the plate surface, branching will be energetically advantageous also when the surface of the plate is perpendicular to an easy axis. Instead of the structure shown in Fig. 10, where $\sin \gamma = H_0/4\pi M$, $\delta_1 = (\pi/4) - (\gamma/2)$, and $\delta_2 = (\pi/4) + (\gamma/2)$, and which was calculated in^[34] (see also^[2]), in the case $H_0 \sim 2\pi M$ the structure shown in Fig. 11 should be formed. All the formulas for this case are analogous to the preceding ones. They are obtained from formulas (2.3)–(2.5) by setting $\gamma = 0$ and making the substitution

$$k \rightarrow k [1 - (H_0/4\pi M)^2].$$

*We have not been able to allow for the numerical multiplier, since the energy of the branched structure in the case under consideration could not be calculated.

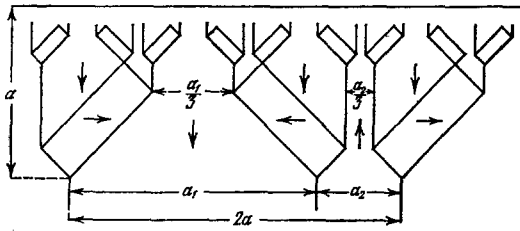


FIG. 11

3. BULK PROPERTIES OF DOMAIN STRUCTURES. THEORY OF THE IDEAL MAGNETIZATION (POLARIZATION) CURVE

a) **Statement of the problem.** In the preceding chapter we saw that a division into domains leads to an energy advantage proportional to the volume of the specimen. The energy due to emergence of the domains at the surface is small. Thus, for example, in the case of the branched structure in a uniaxial ferromagnet this energy is proportional to the 1/3 power of the specimen volume. In many problems the effects due to the emergence of domains at the surface may be unimportant. Such problems include, in particular, the problem of the ideal magnetization (polarization) curve.

The bulk properties of domain structures are simplest in massive ellipsoidal bodies, in plane-parallel plates, and in cylindrical specimens of elliptic cross section. If the external field is uniform, then with neglect of effects due to the emergence of domains at the surface, it may be supposed that there exist in the specimen several uniform phases separated by plane-parallel boundaries. In the simplest cases, the number of phases is two. Each of the phases constitutes a system of domains in contact with domains of another phase. In bulk specimens, the domain width is small in comparison with the dimensions of the specimen.

The average values, over the domain structure, of the magnetic field \mathbf{H} and the magnetic induction \mathbf{B} (or of the electric field \mathbf{E} and the electric induction \mathbf{D}) are uniform within such specimens. These values are connected with the external field \mathbf{H}_0 (or \mathbf{E}_0) by the relations

$$H_{0i} = (\delta_{ik} - n_{ik}) \langle\langle H_k \rangle\rangle + n_{ik} \langle\langle B_k \rangle\rangle, \quad (3.1)$$

$$E_{0i} = (\delta_{ik} - n_{ik}) \langle\langle E_k \rangle\rangle + n_{ik} \langle\langle D_k \rangle\rangle; \quad (3.2)$$

here the double angular brackets indicate an average over the specimen volume, and n_{ik} is the demagnetizing- (depolarizing-) coefficient tensor.

The bulk properties of domain structures are completely described by specification of \mathbf{H}_t and \mathbf{B}_n (or \mathbf{E}_t and \mathbf{D}_n), the two angles that determine the orientation of the separation boundaries, and the concentrations of the phases. These quantities satisfy the three equations (3.1) (or (3.2)) and the condition for coexistence of phases

$$F'_1(\mathbf{H}_t, \mathbf{B}_n) = F'_2(\mathbf{H}_t, \mathbf{B}_n)$$

or

$$F'_1(\mathbf{E}_t, \mathbf{D}_n) = F'_2(\mathbf{E}_t, \mathbf{D}_n)$$

The number of equations is two less than the number of unknowns. Two parameters must be determined from

the minimization condition for the total free energy per unit volume

$$\tilde{F}_{\text{tot}}(\mathbf{H}_0) = V^{-1} \int [\tilde{F}(\mathbf{x}) + (H_0^2/8\pi)] d^3\mathbf{x} = \tilde{F}_{\text{tot}}(\mathbf{H}_0 = 0) - \int_0^{\mathbf{H}_0} \langle\langle \mathbf{M} \rangle\rangle d\mathbf{H}_0,$$

$$F_{\text{tot}}(\mathbf{E}_0) = V^{-1} \int [F(\mathbf{x}) - (E_0^2/8\pi)] d^3\mathbf{x} = F_{\text{tot}}(\mathbf{E}_0 = 0) - \int_0^{\mathbf{E}_0} \langle\langle \mathbf{P} \rangle\rangle d\mathbf{E}_0.$$

We consider first a ferromagnetic specimen in the absence of an external field ($\mathbf{H}_0 = 0$). It is easy to show that if $\mathbf{H}_0 = 0$, that is if the external currents \mathbf{j} vanish, then the free energies

$$\tilde{\mathcal{F}} = -(4\pi)^{-1} \int \left(\int_0^{\mathbf{H}(\mathbf{x})} \mathbf{B} d\mathbf{H} \right) d^3\mathbf{x}$$

and

$$\mathcal{F} = \tilde{\mathcal{F}} + (4\pi)^{-1} \int \mathbf{H}(\mathbf{x}) \mathbf{B}(\mathbf{x}) d^3\mathbf{x}$$

coincide, independently of the nature and geometry of the specimen. In fact, $\mathbf{H} \cdot \mathbf{B} = \text{div} [\mathbf{A} \times \mathbf{H}] + \mathbf{A} \cdot \text{curl} \mathbf{H}$, where \mathbf{A} is the vector potential. The second term in this expression vanishes, since $\mathbf{j} = 0$, and consequently the integral of $\mathbf{H} \cdot \mathbf{B}$ reduces to the integral of a divergence, i.e. to zero.

A similar situation occurs in ferroelectric materials if $\mathbf{E}_0 = 0$, that is if the external charges and charges of conductors vanish. In this case $\mathbf{E} \cdot \mathbf{D} = -\text{div}(\varphi \mathbf{D}) + \varphi \text{div} \mathbf{D}$ (φ is the scalar potential), but $\text{div} \mathbf{D} = 0$, and on conductor surfaces $\varphi = \text{const}$ and $\int \mathbf{D}_n d\mathbf{S} = 0$.

The free energy F in a ferromagnet is $U_{\text{an}} + (H^2/8\pi)$. It is evident that when $\mathbf{H}_0 = 0$, the minimum of the free energy is obtained when in the interior of the specimen $\mathbf{H}_1 = \mathbf{H}_2 = 0$, $\mathbf{M}_1 = -\mathbf{M}_2$ (then $U_{\text{an}} = 0$), and $c_1 = c_2$; that is, we have a special case of conditions (1.27). This result is independent of the model. A similar statement is valid also for ferroelectric materials.

b) **Bulk properties of domain structures. Ideal magnetization curve of uniaxial ferromagnets.** We shall show further that in an arbitrary uniform external field \mathbf{H}_0 , independently of the model, the condition (1.27) for coexistence of phases is the condition for a minimum of the volume part of the total free energy \tilde{F}_{tot} .^[14]

For calculation of \tilde{F}_{tot} it is convenient to use the relation (1.15), which can be expressed in the form

$$\tilde{F}_{\text{tot}} = \langle\langle F \rangle\rangle - (8\pi)^{-1} \langle\langle \mathbf{H} \mathbf{B} \rangle\rangle - (\mathbf{H}_0 \langle\langle \mathbf{M} \rangle\rangle / 2).$$

In order to find the condition for a minimum of \tilde{F}_{tot} , we shall calculate in the linear approximation the change of \tilde{F}_{tot} in a small change of the parameters that describe the properties of the domain structure (this means a small change of all the quantities, including $\langle\langle \mathbf{H}_i \rangle\rangle$, $\langle\langle \mathbf{M}_i \rangle\rangle$, the orientation of the separation boundaries, etc., at constant \mathbf{H}_0). The relation (3.1) is equivalent to the following:

$$H_{0i} = \langle\langle H_i \rangle\rangle + 4\pi n_{ik} \langle\langle M_k \rangle\rangle. \quad (3.3)$$

By use of (3.3) it is easy to show that

$$-\mathbf{H}_0 \delta \langle\langle \mathbf{M} \rangle\rangle / 2 = -\langle\langle \mathbf{H} \rangle\rangle \delta \langle\langle \mathbf{M} \rangle\rangle + 4\pi n_{ik} \langle\langle M_k \rangle\rangle \delta \langle\langle M_i \rangle\rangle.$$

By using the fact that

$$4\pi n_{ik} \delta \langle\langle M_i \rangle\rangle = -\delta \langle\langle H_k \rangle\rangle,$$

it is easy to see that

$$\begin{aligned} -\mathbf{H}_0 \delta \langle\langle \mathbf{M} \rangle\rangle / 2 &= -\langle\langle \mathbf{H} \rangle\rangle \delta \langle\langle \mathbf{M} \rangle\rangle - \langle\langle \mathbf{M} \rangle\rangle \delta \langle\langle \mathbf{H} \rangle\rangle \\ &= (8\pi)^{-1} \langle\langle \mathbf{H} \rangle\rangle \delta \langle\langle \mathbf{B} \rangle\rangle - \langle\langle \mathbf{B} \rangle\rangle \delta \langle\langle \mathbf{H} \rangle\rangle. \end{aligned}$$

It is convenient to transform the mean $\langle\langle \mathbf{H} \cdot \mathbf{B} \rangle\rangle$ as follows:

$$\langle\langle \mathbf{H}\mathbf{B} \rangle\rangle = \langle\langle \mathbf{H}_i \mathbf{B}_i \rangle\rangle + \langle\langle H_n B_n \rangle\rangle = \mathbf{H}_i \langle\langle \mathbf{B}_i \rangle\rangle + \langle\langle H_n \rangle\rangle B_n = \langle\langle \mathbf{H} \rangle\rangle \langle\langle \mathbf{B} \rangle\rangle.$$

Therefore the variation $\delta \tilde{F}_{\text{tot}}$ can be expressed in the form

$$\delta \tilde{F}_{\text{tot}} = \delta \langle\langle F \rangle\rangle - (4\pi)^{-1} \langle\langle \mathbf{H} \rangle\rangle \delta \langle\langle \mathbf{B} \rangle\rangle.$$

The variation $\delta \langle\langle F \rangle\rangle$ has the form

$$\delta \langle\langle F \rangle\rangle = \delta c_1 (F_1 - F_2) + \langle\langle \delta F \rangle\rangle = \delta c_1 (F_1 - F_2) + \frac{1}{4\pi} \langle\langle \mathbf{H} \delta \mathbf{B} \rangle\rangle,$$

where δc_1 is the change of concentration, and the average on the right side is carried out with the phase concentrations of the zeroth approximation. Similarly

$$\delta \langle\langle \mathbf{B} \rangle\rangle = \delta c_1 (\mathbf{B}_1 - \mathbf{B}_2) + \langle\langle \delta \mathbf{B} \rangle\rangle = \delta c_1 (\mathbf{B}_{1t} - \mathbf{B}_{2t}) + \langle\langle \delta \mathbf{B} \rangle\rangle.$$

On using

$$\langle\langle \mathbf{H} \delta \mathbf{B} \rangle\rangle - \langle\langle \mathbf{H} \rangle\rangle \langle\langle \delta \mathbf{B} \rangle\rangle = \langle\langle H_n \delta B_n \rangle\rangle - \langle\langle H_n \rangle\rangle \langle\langle \delta B_n \rangle\rangle = \langle\langle (H_n - \langle\langle H_n \rangle\rangle) \delta B_n \rangle\rangle,$$

we finally get

$$\delta \tilde{F}_{\text{tot}} = \delta c_1 (F'_1 - F'_2) + (4\pi)^{-1} [c_1 (H_{1n} - \langle\langle H_n \rangle\rangle) \delta B_{1n} + c_2 (H_{2n} - \langle\langle H_n \rangle\rangle) \delta B_{2n}].$$

It should be remarked that δB_{1n} and δB_{2n} are the projections of $\delta \mathbf{B}_1$ and $\delta \mathbf{B}_2$ along the normal to the unperturbed separation boundary. Therefore $\delta B_{1n} \neq \delta B_{2n}$. If we do not assume in advance that $F'_1 = F'_2$, then the domain structure has three degrees of freedom, and the variations δc_1 , δB_{n1} , and δB_{n2} are independent. Thus from the condition that the first variation $\delta \tilde{F}_{\text{tot}}$ shall vanish, we get the conditions

$$F'_1 = F'_2, \quad H_{n1} = H_{n2} = \langle\langle H_n \rangle\rangle.$$

Thus the thermodynamic boundary condition $F'_1 = F'_2$ may itself be obtained from the minimization condition for \tilde{F}_{tot} .

Thus by minimization of the volume part of the free energy \tilde{F}_{tot} , we obtain only the one additional condition $H_{n1} = H_{n2}$. The number of relations remains one less than the number of parameters determining the properties of the domain structure; that is, there is a degeneracy in the problem. If the field \mathbf{H} ($\mathbf{H}_1 + \mathbf{H}_2 = \mathbf{H}$, $B_{n1} = B_{n2}$, $\tilde{F}_1 = \tilde{F}_2$) and the phase concentrations c_1 and c_2 are given, then the values of $\langle\langle \mathbf{M} \rangle\rangle$ and of \tilde{F}_{tot} do not change on rotation of the phase-separation boundary about the vector $\mathbf{B}_1 - \mathbf{B}_2$.

Thus the relations (1.27) and (3.1) allow a one-parameter family of structures, possessing the same values of $\langle\langle \mathbf{M} \rangle\rangle$ and of \tilde{F}_{tot} . The unknown parameter determining the orientation of the phase-separation boundary, and likewise the thickness of the layers, can be found only by taking into account effects due to the emergence of the domains at the surface. Nevertheless the dependence of $\langle\langle \mathbf{M} \rangle\rangle$ on \mathbf{H}_0 (the ideal magnetization curve) can be determined as a result of solution of an immensely simpler problem: for given \mathbf{H}_0 , it is necessary only to consider an arbitrary one of the structures satisfying conditions (1.27) and (3.1), and to calculate $\langle\langle \mathbf{M} \rangle\rangle$ for it.

If the number of coexisting phases $N > 2$, but the domains as before are plane-parallel layers,* then the

*Structures of the "checkerboard" type and cylindrical structures have a smaller number of degrees of freedom and can be realized only in the case $N = 2$.

first variation $\delta \tilde{F}_{\text{tot}}$ has the form

$$\delta \tilde{F}_{\text{tot}} = \sum_{\lambda=1}^{\lambda=N} \delta c_{\lambda} F'_{\lambda} + (4\pi)^{-1} \langle\langle (H_n - \langle\langle H_n \rangle\rangle) \delta B_n \rangle\rangle.$$

For $N = 3$, the conditions $H_{n1} = H_{n2} = H_{n3}$ together with the conditions for phase coexistence (1.21) and (1.23) and the relations (3.1) uniquely determine the bulk properties of the domain structure. Coexistence of three phases is possible, for example, in cubic ferromagnets if the internal field \mathbf{H} is parallel to the diagonal axis [111] and the separation boundaries are perpendicular to the field.

Coexistence of four phases is possible, it appears, only in special cases. In particular, in cubic ferromagnets four phases can coexist if the field \mathbf{H} is parallel to one of the easy axes and the phase-separation boundaries are perpendicular to the field. If in a hexagonal ferromagnet the uniaxial-anisotropy constant β is negative (cobalt has this property at temperatures above 200°C), then in a field \mathbf{H} parallel to the hexagonal axis, coexistence of six phases is possible.

Since formulas (1.14) for \mathcal{F}_{tot} in dielectrics are analogous to formulas (1.15) for $\tilde{\mathcal{F}}_{\text{tot}}$ in magnets, it is obvious that in the interior of a ferroelectric specimen the phase-coexistence conditions (1.28) must be satisfied.

In a uniaxial ferromagnet the conditions $\mathbf{H} = \text{const}$, $B_n = \text{const}$, $\tilde{F} = \text{const}$, as has already been pointed out, mean that the phase-separation boundary is parallel to the easy axis and the field \mathbf{H} is perpendicular to this axis. For given phase concentrations c_1 and c_2 ($c_1 + c_2 = 1$) and for given field $\mathbf{H} \perp z$, the volume part of the magnetization $\langle\langle \mathbf{M} \rangle\rangle$, as well as \tilde{F}_{tot} , remains unchanged on rotation of the separation boundary about the z axis. This makes it possible to find the relation between $\langle\langle \mathbf{M} \rangle\rangle$ and \mathbf{H}_0 in the range where a domain structure exists, for all orientations of \mathbf{H}_0 and of the crystallographic axes with respect to the ellipsoid axes.^[35]

We shall take into account that for $H_z = 0$ and $H_x^2 + H_y^2 < (\beta M)^2$ the "equation of state" of a uniaxial ferromagnet takes the form

$$\beta M_x = H_x, \quad \beta M_y = H_y, \quad M_z = \pm (M^2 - M_x^2 - M_y^2)^{1/2}.$$

Since $\mathbf{H} = \text{const}$, the relation (3.3) can be rewritten in the form

$$H_{0i} = H_i + 4\pi n_{ik} \langle\langle M_k \rangle\rangle,$$

or, further, in the form

$$H_{0i} = 4\pi \tilde{n}_{ik} \langle\langle M_k \rangle\rangle, \quad (3.4)$$

where

$$\tilde{n}_{ik} = n_{ik} + (\beta/4\pi) (\delta_{ik} - \delta_{i3}\delta_{k3}). \quad (3.5)$$

Formulas (3.4) and (3.5) determine the ideal magnetization curve. The phase concentrations are determined from the conditions

$$\langle\langle M_z \rangle\rangle = (c_1 - c_2) (M^2 - M_x^2 - M_y^2)^{1/2}, \quad M_x = M_{x1} = M_{x2} = \langle\langle M_x \rangle\rangle, \\ M_y = M_{y1} = M_{y2} = \langle\langle M_y \rangle\rangle.$$

c) **Magnetostriction and electrostriction.** So far we have disregarded the energy of elastic deformations. In ferromagnets the magnetostrictive energy is small in comparison with the anisotropy energy and the magnetostatic energy, but there are materials in which they are comparable (see, for example, the book^[36]). In ferro-

electric materials the electrostrictive energy is, as a rule, comparable with the other forms of energy.^[5] We note also that the problem of elastic deformations during magnetization (polarization) is of independent interest. We shall here present briefly some results obtained in^[35].

Magnetostrictive effects are described by the thermodynamic relation for the free energy \tilde{F} (see Appendix)

$$d\tilde{F} = (\sigma_{ik} - \tilde{F}\delta_{ik} - \frac{1}{4\pi} H_i B_k) d\frac{\partial u_i}{\partial x_k} - \frac{1}{4\pi} B dH, \quad (3.6)$$

where $\sigma_{ik} = \sigma_{ki}$ is the stress tensor. In an anisotropic body, the free energy depends not only on the symmetric components of the deformation tensor, $u_{ik} = (1/2) \times [(\partial u_i / \partial x_k) + (\partial u_k / \partial x_i)]$, but also on the antisymmetric, $v_{ik} = (1/2) [(\partial u_i / \partial x_k) - (\partial u_k / \partial x_i)]$.^{*} We shall introduce also the free energy F' in the variables H_t , B_n , and $\partial u_i / \partial x_k$, where the indices t and n designate the components tangential and normal to the phase-separation boundary:

$$F' = \tilde{F} + (4\pi)^{-1} H_n B_n,$$

$$dF' = (\sigma_{ik} - F'\delta_{ik} + \frac{1}{4\pi} H_n B_n \delta_{ik} - \frac{1}{4\pi} H_i B_k) d\frac{\partial u_i}{\partial x_k} - \frac{1}{4\pi} B_t dH_t + \frac{1}{4\pi} H_n dB_n.$$

Instead of the quantities \tilde{F} and F' , which refer to unit volume of the material, it is convenient in the theory of elasticity to introduce the free energy corresponding to a given mass, and specifically to the mass of unit undeformed volume. To such quantities we shall attach the subscript 0:

$$d\tilde{F}_0 = (\sigma_{ik} - \frac{1}{4\pi} H_i B_k) d\frac{\partial u_i}{\partial x_k} - \frac{1}{4\pi} (1 + u_{ii}) B dH,$$

$$dF'_0 = (\sigma_{ik} + \frac{1}{4\pi} H_n B_n \delta_{ik} - \frac{1}{4\pi} H_i B_k) d\frac{\partial u_i}{\partial x_k} + \frac{1}{4\pi} (1 + u_{ii}) (-B_t dH_t + H_n dB_n).$$

On the phase-separation boundaries, besides H_t and B_n , the values of $\partial u_i / \partial x_\alpha$ and of $\sigma_{ik} n_k = \sigma_{in}$ are also conserved; here the index α numbers the components in the plane of the boundary. In order to obtain the condition for coexistence of phases, it is necessary to construct the thermodynamic potential in the conserved variables. For this purpose we put dF'_0 into the form

$$dF'_0 = (\sigma_{i\alpha} + \frac{1}{4\pi} H_n B_n \delta_{i\alpha} - \frac{1}{4\pi} H_i B_\alpha) d\frac{\partial u_i}{\partial x_\alpha} + (\sigma_{\alpha n} - \frac{1}{4\pi} H_\alpha B_n) d\frac{\partial u_\alpha}{\partial n} + \sigma_{nn} d\frac{\partial u_n}{\partial n} + \frac{1}{4\pi} (1 + u_{ii}) (-B_t dH_t + H_n dB_n).$$

The desired thermodynamic potential, which has a minimum for given H_α , B_n , $\partial u_i / \partial x_\alpha$, and σ_{in} , is the thermodynamic potential

$$\Phi'_0(H_\alpha, B_n, \frac{\partial u_i}{\partial x_\alpha}, \sigma_{\alpha n} - \frac{1}{4\pi} H_\alpha B_n, \sigma_{nn}) = F'_0 - (\sigma_{\alpha n} - \frac{1}{4\pi} H_\alpha B_n) \frac{\partial u_\alpha}{\partial n} - \sigma_{nn} \frac{\partial u_n}{\partial n}.$$

The complete system of boundary conditions has the form

$$H_\alpha = \text{const}, \quad B_n = \text{const}, \quad \frac{\partial u_i}{\partial x_\alpha} = \text{const}, \quad \sigma_{in} = \text{const},$$

$$\Phi'_0(H_\alpha, B_n, \frac{\partial u_i}{\partial x_\alpha}, \sigma_{\alpha n} - \frac{1}{4\pi} H_\alpha B_n, \sigma_{nn}) = \text{const}. \quad (3.7)$$

We shall now consider an ellipsoidal specimen, whose shape is maintained constant, placed in a uniform external field H_0 . In such a specimen there is the possibility of a nonuniform deformation (changing from domain to domain), which on the average is zero.

^{*}In the literature it is often asserted that in an anisotropic body the stress tensor σ_{ik} is asymmetric (see, for example, the books [37]). This assertion is incorrect. The correct result is obtained by taking account of the asymmetric (proportional to v_{ik}) terms in the expression for the free energy.

The bulk properties of the domain structure are completely determined by specification of the eight parameters that do not change on crossing a separation boundary,

$$H_\alpha, B_n, \sigma_{in}, n,$$

and of the concentration c'_i (by weight) of one of the two phases (n is the unit vector normal to the plane of the phase-separation boundary). Another six such parameters ($\partial u_i / \partial x_\alpha$) are zero, since the ellipsoid remains undeformed on the average. These quantities satisfy the three equations (3.1), the phase-coexistence condition (3.7), and the three additional equations

$$\left\langle \frac{\partial u_i}{\partial n} \right\rangle = 0, \quad (3.8)$$

where

$$\langle f \rangle = c'_1 f_1 + c'_2 f_2.$$

Thus the number of equations is seven, two less than the number of parameters that determine the bulk properties of the domain structure. In addition, it is necessary to obtain the conditions for minimization of the volume part of the total free energy \tilde{F}_t , considered as a function of the nine parameters that determine the bulk properties of the domain structure, under the seven supplementary conditions (3.1), (3.7), and (3.8).

In^[35] it was shown that from the condition for a minimum of \tilde{F}_t there is obtained only one additional condition $H_{n1} = H_{n2}$, just as in the absence of magnetostriction. Thus the degeneracy in the problem is retained. But in the presence of magnetostriction, this degeneracy can no longer be interpreted as simply as before.

To this point we were dealing with an ellipsoid whose shape was assumed to remain unchanged. This means that to the body are applied mechanical forces that prevent a change of its shape. If such forces are absent, then a body that has the form of an ellipsoid in the demagnetized (unpolarized) state can deform on application of an external field and, in particular, can turn in the external field under the action of the purely magnetostatic Maxwell stresses

$$\sigma_{ik} = (4\pi)^{-1} [H_i H_k - (H^2 \delta_{ik} / 2)].$$

The torque can be absent only in definite cases, for example in the cases of a long cylinder (wire) or plane-parallel plate in an external field H_0 parallel to it. It makes sense to consider just such cases. The ellipsoidality of the specimen is conserved in such cases except for effects due to the emergence of domains at the surface and unimportant in the calculation of the volume energy. This is due to the fact that the mean deformation $\partial u_i / \partial x_k$ will be uniform. The same situation can occur also in cases in which the body is subjected to applied stresses that do not produce a torque. Such a situation occurs, for example, in experiments with stretched wires. In order to investigate the bulk properties of the domain structures in these cases, we point out that in the previous problem (an ellipsoid of fixed shape), the concept of the undeformed state had a conditional character, since it was not assumed that in the absence of deformation ($\partial u_i / \partial x_k = 0$) and in the absence of field ($H = 0$) the stresses σ_{ik} were also zero. We shall now define the undeformed state in this same way.

Then it is obvious that in those cases in which the ellipsoidality of the specimen is conserved (although the ratios of the semiaxes may change) and the specimen remains uniform on the average, in the interior of the specimen the following relations must be satisfied:

$$\mathbf{H} = \text{const}, B_n = \text{const}, \Phi'_0 = \text{const}, \partial u_i / \partial x_\alpha = \text{const}, \sigma_{in} = \text{const}, \langle v_{ih} \rangle = 0.$$

The last relation is the condition that the body as a whole shall not turn in the external field.

In^[35] the magnetostriction of a uniaxial ferromagnet was calculated with allowance for the domain structure. Analogous results are valid also for ferroelectric materials (in all formulas of this section it is necessary only to make the substitution $\mathbf{H} \rightarrow \mathbf{E}$, $\mathbf{B} \rightarrow \mathbf{D}$).

4. THEORY OF FINE FERROMAGNETIC PARTICLES

a) **Brown's theorem.**^[36] So far, we have considered massive specimens, whose dimensions were assumed to be large in comparison with the thickness δ of a domain wall. If the dimensions of the specimen are less than δ , then the specimen should be single-domain. This was first predicted by Frenkel and Dorfman.^[39] For simplicity, we shall hereafter consider spherical ferromagnetic particles with uniaxial anisotropy, in the absence of an external field. Brown showed that if the radius R of such a particle is less than a certain critical value R_c , the magnetization \mathbf{M} in the specimen will be rigorously uniform.* The possibility of a uniform state explains the large values of the coercive force of fine particles in the case of weak anisotropy.

The free energy $\mathcal{F} = \mathcal{F}$ of a ferromagnetic particle can be expressed as the sum of three terms

$$\mathcal{F}_{\text{nonunif}} = (1/2)\alpha \int (\partial M_i / \partial x_k)^2 dV, \\ \mathcal{F}_{\text{an}} = (1/2)\beta \int (M_x^2 + M_y^2) dV,$$

$$\mathcal{F}_m = \int (H^2/8\pi) d^3x = \int [(H^2/8\pi) - (\mathbf{H}\mathbf{B}/8\pi)] d^3x = -\langle \mathbf{H}\mathbf{B} \rangle \int d^3x.$$

If the magnetization in the specimen is uniform and directed along the axis of easy magnetization, the free energy of the particle is

$$\mathcal{F}_m \geq (2\pi \langle \mathbf{M} \rangle^2 / 3) V. \tag{4.1}$$

We shall now show that in a nonuniform state with mean magnetization $\langle \mathbf{M} \rangle$

$$\mathcal{F}_{\text{unif}} = (2\pi M^2 / 3) V. \tag{4.2}$$

To this end we consider the field $\mathbf{H}_{\langle \mathbf{M} \rangle}(\mathbf{x})$ produced by a spherical particle with uniform magnetization $\langle \mathbf{M} \rangle$ (inside the specimen $\mathbf{H}_{\langle \mathbf{M} \rangle} = -4\pi \langle \mathbf{M} \rangle / 3$) and the field $\delta \mathbf{H} = \mathbf{H} - \mathbf{H}_{\langle \mathbf{M} \rangle}$, where \mathbf{H} is the true field produced by the nonuniform magnetization of the particle. Then

$$\mathcal{F}_m = \int (H^2/8\pi) d^3x \geq \int [(H_{\langle \mathbf{M} \rangle}^2/8\pi) + (\mathbf{H}_{\langle \mathbf{M} \rangle} \delta \mathbf{H}/4\pi)] d^3x \\ = (2\pi \langle \mathbf{M} \rangle^2 / 3) V + \int [\mathbf{H}_{\langle \mathbf{M} \rangle} \delta \mathbf{B} - 4\pi \delta \mathbf{M}] / 4\pi d^3x.$$

We note further that

$$\int (\mathbf{H}_{\langle \mathbf{M} \rangle} \delta \mathbf{B} / 4\pi) d^3x = 0,$$

*We emphasize that in massive spherical specimen with small anisotropy, the state with uniform magnetization would be absolutely unstable, since the demagnetizing field $\mathbf{H} = -4\pi \mathbf{M} / 3$ would be beyond the limits of the metastability range (see paragraph b of Chapter 1).

since $\mathbf{H}_{\langle \mathbf{M} \rangle} = -\nabla \phi_{\langle \mathbf{M} \rangle}$ and $\delta \mathbf{B} = \text{curl } \delta \mathbf{A}$, and that

$$\int \mathbf{H}_{\langle \mathbf{M} \rangle} \delta \mathbf{M} d^3x = 0,$$

since inside the specimen $\mathbf{H}_{\langle \mathbf{M} \rangle}$ is uniform and $\langle \delta \mathbf{M} \rangle = 0$. This proves the inequality (4.2).

Thus it can be stated that

$$\mathcal{F} \geq \min \mathcal{F}_0, \quad \mathcal{F}_0 = \int \left[\frac{\alpha}{2} \left(\frac{\partial M_i}{\partial x_k} \right)^2 + \frac{\beta}{2} (M_x^2 + M_y^2) + \frac{2\pi}{3} \langle \langle M_i \rangle \rangle M_i \right] dV.$$

Instead of minimizing the right side under the condition $M_i^2 = M^2$, we shall weaken this condition, requiring only that

$$V^{-1} \int M_i^2 dV = M^2. \tag{4.3}$$

In addition, we shall require also that on the boundaries of the specimen the condition $\partial M_i / \partial n = 0$ shall be satisfied, as it must be by the exact solution of the nonuniform problem (see Sec. b of Chapter 1).

On varying \mathcal{F}_0 under the condition (4.3), we get the linear equation

$$-\alpha \Delta M_i + \beta M_i (1 - \delta_{i3}) + (4\pi/3) \langle \langle M_i \rangle \rangle = \lambda M_i, \tag{4.4}$$

where the characteristic quantity λ is a Lagrange multiplier. The desired minimum can be expressed as follows in terms of λ min:

$$\min \mathcal{F}_0 = \lambda_{\text{min}} M^2 V / 2. \tag{4.5}$$

Equation (4.4) has a uniform solution with $\mathcal{F}_0 = \mathcal{F}_{\text{unif}}$ (see formula (4.1)). In the nonuniform case the minimal λ corresponds to a solution of the form

$$M_z = \text{const} \cdot j_1(kr) Y_{1m}(n), \quad M_x = M_y = 0, \quad k^2 = \lambda / \alpha,$$

where $j_1(x) = I_{3/2}(x) / x^{1/2}$, and $I_{3/2}$ is a Bessel function. The eigenvalue of λ is found from the boundary condition $\partial M_i / \partial n = 0$, which in this case takes the form

$$j_1'(kR) = 0,$$

that is,

$$kR = x_0 = 2.08, \quad \lambda = \alpha x_0^2 / R^2,$$

where x_0 is the first zero of the function $j_1'(x)$.

With use of (4.5) it is easy to see that the uniform state is with certainty advantageous if

$$R < (3\alpha/4\pi)^{1/2} x_0.$$

Thus we have proved the existence of a critical radius and have found a lower bound to it.

It is obvious that in the case of high anisotropy ($\beta/4\pi \gg 1$) the critical radius $R_c \gg \delta \sim (\alpha/\beta)^{1/2}$. It is natural to expect that in this case the uniform state ceases to be advantageous when a splitting into domains becomes advantageous. By comparison of the free energy of a two-domain structure (Fig. 12) with $\mathcal{F}_{\text{unif}}$, it is possible to find an upper bound to R_c . The free energy of the structure shown in Fig. 12 is

$$\mathcal{F} = (8\pi^2 \gamma t^2 M^2 / \delta) + 2(\alpha\beta)^{1/2} M^2 \pi R^2, \quad \gamma = 0.245$$

(the first term is the magnetostatic energy, the second

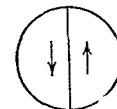


FIG. 12

is the surface-tension energy on the phase-separation boundary). After simple calculations, we obtain finally*

$$(3\alpha/4\pi)^{1/2}x_0 \leq R_c \leq 3(\alpha\beta)^{1/2}/4\pi^2 [(4/3)-2\gamma] \quad (\beta/4\pi \gg 1).$$

We note that $(\alpha\beta)^{1/2} \sim \beta\delta$. Apparently in the case of high anisotropy our upper bound coincides, at least in order of magnitude, with the true value of the critical radius R_c .

b) **Critical radius in the case of small anisotropy.** In the case of small anisotropy it can be shown, on the basis of the work of Frei et al.,^[40] that the upper bound to the critical radius R_c closes in on the lower, so that the formula

$$R_c = (3\alpha/4\pi)^{1/2}x_0 \quad (4.6)$$

becomes exact in the limit $\beta/4\pi \rightarrow 0$.

Frei et al.^[40] considered the stability of the uniform state of a spherical particle with respect to infinitely small perturbations; this offers the possibility of finding an upper bound to R_c . It was assumed that the components of the magnetization in the nonuniform state have the form

$$M_z = M \cos \omega \approx M [1 - (1/2)\omega^2], \quad M_\varphi = M \sin \omega \approx M\omega, \\ \omega = \omega(r, \theta) \ll 1,$$

where r and θ are spherical coordinates and M_φ is the azimuthal projection of the vector \mathbf{M} .

The change of field $\delta\mathbf{H}(\mathbf{x}) = \mathbf{H}(\mathbf{x}) - \mathbf{H}_0(\mathbf{x})$ is proportional to the change of the density of fictitious magnetic charges. The latter in turn is proportional to ω^2 .

Therefore the change in the magnetostatic energy, $\delta\mathcal{F}_m$, is

$$\delta\mathcal{F}_m = \int (\mathbf{H}_0 \delta\mathbf{H}/4\pi) d^3\mathbf{x} = \int [\mathbf{H}_0 (\delta\mathbf{B} - 4\pi\delta\mathbf{M})/4\pi] d^3\mathbf{x} = \\ = - \int \mathbf{H}_0 \delta\mathbf{M} dV = - (2\pi M^2/3) \int \omega^2 dV.$$

Neglecting the anisotropy energy, we write the change of the total free energy as follows:

$$\delta\mathcal{F} = \pi M^2 \int_0^R r^2 dr \int_0^\pi \sin \theta d\theta \left\{ \alpha \left[\left(\frac{\partial \omega}{\partial r} \right)^2 + \frac{1}{r^2} \left(\frac{\partial \omega}{\partial \theta} \right)^2 + \frac{\omega^2}{r^2 \sin^2 \theta} \right] - \frac{4\pi\omega^2}{3} \right\}.$$

We separate $\delta\mathcal{F}$ into two terms:

$$\delta\mathcal{F} = \delta\mathcal{F}_1 + \delta\mathcal{F}_2,$$

$$\delta\mathcal{F}_1 = \pi\alpha M^2 \int_0^R r^2 dr \int_0^\pi \sin \theta d\theta \left[\left(\frac{\partial \omega}{\partial r} \right)^2 + \frac{1}{r^2} \left(\frac{\partial \omega}{\partial \theta} \right)^2 + \frac{\omega^2}{r^2 \sin^2 \theta} - \frac{x_0^2 \omega^2}{R^2} \right],$$

$$\delta\mathcal{F}_2 = \pi\alpha M^2 x_0^2 (R^2 - R_c^2) \int_0^R r^2 dr \int_0^\pi \sin \theta d\theta \cdot \omega^2,$$

here R_c is determined by formula (4.6), and x_0 was defined in the preceding section.

The condition for a minimum of $\delta\mathcal{F}_1$ has the form

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \omega}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial \omega}{\partial \theta} \right) - \frac{\omega}{r^2 \sin^2 \theta} + k^2 \omega^2 = 0, \quad (4.7) \\ \left(\frac{\partial \omega}{\partial r} \right)_{r=R} = 0, \quad k^2 = \frac{x_0^2}{R^2}.$$

If one multiplies this expression by ω and integrates over the volume, it is easy to verify that the minimum of $\delta\mathcal{F}_1$ is zero.

The solution of the boundary problem (4.7) is

$$\omega \sim j_1(kr) \sin \theta.$$

Since for such a perturbation $\delta\mathcal{F}_2 = 0$, it is easy to see

*We have here somewhat improved the estimate from above obtained by Brown.^[38]

that when $R > R_c$ the value of $\delta\mathcal{F}$ is negative. On taking account of the result obtained in the previous section, we conclude that formula (4.6) determines the exact value of the critical radius.

5. THEORY OF NUCLEI OF REVERSE MAGNETIZATION*

a) **Thermal activation of nuclei.** As is well known, the reason for magnetic hysteresis is the possibility of existence of metastable states. In a uniaxial ferromagnet, the metastable states can be realized in case

$$H_z^{2/3} + (H_x^2 + H_y^2)^{1/3} < (\beta M)^{2/3}.$$

If the field \mathbf{H} is directed along the axis of easy magnetization ($H_x = H_y = 0$) and is less in absolute value than βM , the state with magnetization antiparallel to the field is metastable.

The metastable state can be destroyed if, for example, as a result of thermal fluctuations there is formed a nucleus of opposite magnetization whose dimensions are sufficiently large so that growth of the nucleus will lead to a diminution of the free energy \mathcal{F} of the body.^[42] We recall that a metastable state is stable with respect to infinitely small perturbations (such perturbations increase the free energy of the body), and that consequently, for formation of a nucleus that can grow, it is necessary to surmount a finite energy barrier. The probability of formation of such nuclei as a result of thermal fluctuations is proportional to $\exp(-R_{\min}/T)$, where T is the temperature; the energy barrier R_{\min} is the work necessary for formation of a so-called critical nucleus, which is in unstable equilibrium with its surroundings: its shape is such that for given thickness of the nucleus, the free energy \mathcal{F} of the body will be a minimum, and the thickness corresponds to a maximum of the free energy $\tilde{\mathcal{F}}$. Thus the critical nucleus corresponds to a saddle point of the functional $\tilde{\mathcal{F}}z(x, y)$, where the function $z(x, y)$ describes the form of the nucleus. Here we assume that the dimensions of the nucleus are large in comparison with the domain-wall thickness δ ; this, as will be shown below, is correct only in the case of weak metastability ($H \ll \beta M$). It is only in this case that the nuclei can be treated within the framework of macrotheory; in the contrary case, the parameters of the critical nucleus must be found as the result of a solution of the static equation of Landau and Lifshitz,^[13] $\mathbf{M} \times \mathbf{H}_{\text{eff}} = 0$.

In the work of Döring^[43] (see also^[2]), only nuclei of ellipsoidal form were considered, and the possibility of a deviation of the magnetization from the easy axis was disregarded. In the weak-anisotropy case, this deviation leads to a significant decrease in the sum of the magnetostatic energy and the magnetic anisotropy energy. In reference^[41] the exact form of the critical nuclei was obtained for a number of limiting cases. It was shown that the energy barrier R_{\min} in the weak-anisotropy case is significantly smaller than in Döring's.

The work R that we must calculate is equal to the change of the free energy \mathcal{F} due to the presence of the nucleus. It can be expressed in the form (see^[41])

$$R = \int [-\delta\mathbf{M} \cdot \mathbf{H} + \{(\delta\mathbf{H})^2/8\pi\} + U_{\text{an}}] d^3\mathbf{x} + \int \Delta dS. \quad (5.1)$$

*In this chapter we quote results obtained in^[41].

In formula (5.1), \mathbf{H} is the field in the uniform metastable state (that is, far from the nucleus), and $\delta\mathbf{H}$ and $\delta\mathbf{M}$ are the changes of field and of magnetization due to the formation of the nucleus. The last term is the surface tension on the phase-separation boundary, and the integration extends over the surface of the nucleus.

We consider the surface tension. In Chapter 1 it was shown that for boundaries inclined with respect to the axis of easy magnetization, the concept of surface tension when $\beta/4\pi \ll 1$ has meaning only for small angles of inclination of the boundary. It can be shown^[41] that in the case $\beta/4\pi \gg 1$, in fields $H \ll \beta M$, the surface tension can be defined with good accuracy for an arbitrary inclination of the boundary and is independent of the angle of inclination. Below, we must consider only such boundaries, for which the surface tension Δ can be defined in accordance with (1.34).

In closing this section, we note that on the boundaries of the nucleus the thermodynamic boundary condition (1.23) is not satisfied. The reasons for this are the same as those for which, in the analogous problem of nuclei in a liquid-vapor system, the condition of equality of pressure is not satisfied.

b) **Elongated nuclei (weak fields).** In the case to be considered (uniaxial ferromagnet; $\mathbf{H} \parallel z$), the critical nucleus is symmetric about the easy axis (the z axis). An axial section of the nucleus is shown schematically in Fig. 13. The form of the critical nucleus is described by the function $\rho_0(z)$.

In a field small in comparison with βM , the "equation of state" of a uniaxial ferromagnet has the form

$$\delta H_\rho = \beta M_\rho,$$

that is, the magnetic permeability μ in a direction perpendicular to the easy axis is

$$\mu = 1 + 4\pi\beta^{-1}.$$

It will be shown below that in a weak field ($\mu H \ll 4\pi M$) the nucleus is elongated along the easy axis ($\rho_0(0) \ll l$; see Fig. 13) and the deviation of the magnetization from the easy axis is small ($M_\rho \ll M$). In this case the equation $\text{div } \mathbf{B} = 0$ can be linearized:

$$\text{div } \mathbf{B} = \frac{\mu}{\rho} \frac{\partial}{\partial \rho} (\rho \delta H_\rho) + \frac{\partial \delta H_z}{\partial z} = 0.$$

The field $\delta\mathbf{H}$ and the polarization M_ρ are produced by fictitious "magnetic charges," concentrated on the surface of the nucleus. Since the nucleus is elongated, the "charge" density decreases slowly, and at points not too far from the nucleus $\delta H_z \ll \mu \delta H_\rho$ (in the case of an infinite, uniformly charged cylinder, the field δH_z would be zero). Inside the nucleus the field δH_ρ is also small (appreciably smaller than outside it). Therefore in the first approximation with respect to $\rho_0(0)/l$, the magne-

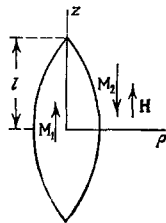


FIG. 13

tization M_l inside the nucleus does not deviate from the axis of easy magnetization:

$$M_\rho^{(1)} = \delta H_\rho^{(1)} = 0$$

On the phase-separation boundary (at $\rho = \rho_0(z)$), the condition $B_n = \text{const}$ must be fulfilled:

$$-4\pi M \dot{\rho}_0 = 4\pi M \dot{\rho}_0 + \mu \delta H_\rho^{(2)} \quad (\dot{\rho}_0 = d\rho_0/dz),$$

that is,

$$\mu \delta H_\rho^{(2)} = -8\pi M \dot{\rho}_0.$$

At not too great distances, the field $\delta H_\rho^{(2)}$ is

$$\delta H_\rho^{(2)} = -8\pi M \dot{\rho}_0 / \mu \rho. \quad (5.2)$$

In order to determine the range of applicability of this formula, we make in the equations of magnetostatics the substitution

$$\delta H_\rho = M h_\rho, \quad \delta H_z = \mu^{1/2} M h_z, \quad \rho = \mu^{1/2} r.$$

Then the equations of magnetostatics take the form

$$\text{div } \mathbf{h} = 0, \quad \text{rot } \mathbf{h} = 0,$$

where the field \mathbf{h} is produced by surface "charges"

$$\sigma(z) = -(2/\mu) \dot{\rho}_0(z).$$

For $r \gg l$ the field \mathbf{h} has the dipole form; that is, it decreases as $(r^2 + z^2)^{-3/2}$. It is obvious that formula (5.2) is valid only when

$$\rho \ll L \sim \mu^{1/2} l.$$

In this range, the sum of the anisotropy energy and the energy of the demagnetizing field $\delta\mathbf{H}$ is

$$(1/2) \beta M_\rho^2 + [(\delta\mathbf{H})^2/8\pi] \approx (\delta H_\rho)^2/8\pi \gg (\delta H_z)^2/8\pi.$$

On truncating the logarithmic integral (over ρ) at distances $\rho \sim \mu^{1/2} l$, we get

$$R = \int_{-l}^l \{ \mu^{-1} (4\pi M)^2 \ln [\mu^{1/2} l / \rho_0(0)] \rho_0^2 \dot{\rho}_0^2 + 2\pi M H \rho_0 (\rho_m - \rho_0) \} dz;$$

here we have introduced the notation

$$\rho_m = \Delta/MH = 2\beta\delta M/H \gg \delta.$$

The saddle point of the functional R , that is the critical nucleus, corresponds to a shape given by the equation (see^[41])

$$z/\rho_m = \pm [(4\pi M/\mu H) \ln (4\pi M/\mu H)]^{1/2} \{ \arcsin (1 - \rho_0/\rho_m^{-1}) + \rho_m^{-1} [\rho_0 (\rho_m - \rho_0)]^{1/2} \}.$$

The energy barrier R_{\min} is

$$R_{\min} = (\mu^2/4) \beta^2 M^2 \delta^3 (4\pi M/\mu H)^{3/2} [\ln (4\pi M/\mu H)]^{1/2}.$$

c) **Spherical nuclei in the case of strong anisotropy, and surface nuclei.** 1) **Spherical nuclei.** In the case $\beta/4\pi \lesssim 1$, the range $\mu H \ll 4\pi M$ coincides with the range of weak metastability, $H \ll \beta M$. In the case of strong anisotropy ($\beta/4\pi \gg 1$), the case $4\pi M \ll H \ll \beta M$ is also possible; it will be investigated below.

If the anisotropy is large, then inclination of the magnetization to the easy axis is energetically disadvantageous ($U_{\text{an}} = 0$). On the boundary of the nucleus, the normal σ component of the magnetization undergoes a jump $\sigma = 2M_{1n}$, which can be considered as a surface density of fictitious magnetic charges, producing a demagnetizing field $\delta\mathbf{H}$.

In the case under consideration, l turns out to be of order $\rho_0(0)$, and the role of the demagnetizing field is unimportant:

$$(\delta H)^2/8\pi \sim 4\pi M^2 \ll MH.$$

The work R can be expressed in the form

$$R = -2MHV + \Delta S = MH(2V + \rho_m S), \quad (5.3)$$

where V and S are the volume and surface of the nucleus. This expression differs only by the multiplier of V from the corresponding expression for an isotropic liquid-vapor system. It is therefore obvious that the critical nucleus must be spherical; that is,

$$R = 4\pi MH [-(2r^3/3) + \rho_m r^2],$$

where r is the radius of the nucleus. The maximum of the function $R(r)$ corresponds to

$$r = \rho_m, \\ R_{\min} = (4\pi/3) MH \rho_m^3 = (32\pi/3) \beta M^2 \delta^3 (\beta M/H)^2.$$

2) Surface nuclei. In certain cases the formation of surface nuclei may be more probable, since the corresponding energy barrier $R_{\min}^{(S)}$ is smaller than the barrier that determines the probability of formation of nuclei within the volume of the specimen (the latter we shall here denote by $R_{\min}^{(V)}$).*

The simplest example is the case $4\pi M \ll H \ll \beta M$. We comment here that the field $\mathbf{H} = \{0, 0, H\}$ is the internal field, which does not coincide with the field $\mathbf{H}^{(e)}$ in a vacuum. The latter can be determined from the conditions of continuity of \mathbf{H}_t and \mathbf{B}_n on the surface of the specimen. The work R is determined by formula (5.3), where V is the volume of the nucleus and S is the area of the separation boundary. On setting $V = V'/2$ and $S = S'/2$, where V' and S' are the volume and the surface area of the doubled figure, it is easy to conclude that the critical nucleus has the form of a hemisphere and that

$$R_{\min}^{(S)} = R_{\min}^{(V)}/2 = (16\pi/3) \beta M^2 \delta^3 (\beta M/H)^2.$$

We emphasize that this result is independent of the orientation of the easy axis with respect to the surface of the specimen.

A representative value of R_{\min}/T for typical ferromagnets (Fe, Co) at room temperature is of order 10^3 , so that under ordinary conditions thermal activation of nuclei is impossible. But for reasonable proximity to the Curie temperature, the barrier R_{\min} may be significantly decreased in consequence of the decrease of magnetization (the values of β and δ change little on approach to the Curie point). The formulas obtained are still applicable near the Curie temperature, where thermal activation of nuclei should become observable. Our estimates permit us to conjecture that the barrier R_{\min} will be small in an observable region near the limit of metastability (for $H = \beta M$, the barrier $R_{\min} = 0$).

Recent papers of Lifshitz and Kagan^[44] and of Iordanskiĭ and Finkel'shtein^[45] investigated the formation of nuclei as a result of quantum-mechanical tunnel-

ing. In this case the probability of formation of nuclei is proportional to $\exp(-2\hbar^{-1} \text{Im } S)$, where $\text{Im } S$ is the imaginary part of the classical action. In the case of a ferromagnet, it is possible to obtain an estimate of the argument of the exponential function by means of the Landau-Lifshitz equation^[13]

$$\frac{d\mathbf{M}}{dt} = \gamma [\mathbf{M}, \mathbf{H}_{\text{eff}}].$$

From dimensional considerations it follows that

$$\hbar^{-1} \text{Im } S = (M\delta^3/\hbar\gamma) f(\beta/4\pi, \beta M/H),$$

where f is an unknown function, whose value for $\beta \sim 4\pi$ and $\beta M - H \sim H \sim \beta M$ should be of order unity. For $M = 10^3$ G and $\delta \sim 10^{-6}$ cm the value is

$$M\delta^3/\hbar\gamma \sim 10^5,$$

and consequently the probability of quantum tunneling is extremely small; this is due to the large value of the "mass" of a domain boundary.^[11,46]

Analogous results for superconductors and for non-ferromagnetic metals were obtained in^[47].

APPENDIX

THERMODYNAMIC THEORY OF MAGNETOSTRICTION. THE STRESS TENSOR

We first find the dependence of the free energy on the antisymmetric components of the deformation tensor

$$v_{ik} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_k} - \frac{\partial u_k}{\partial x_i} \right).$$

We consider, for example, the free energy of a magnet

$$\tilde{F}(\mathbf{H}, \frac{\partial u_i}{\partial x_k}) = \tilde{F}(\mathbf{H}=0, \frac{\partial u_i}{\partial x_k}) - \frac{1}{4\pi} \int_0^H \mathbf{B} d\mathbf{H}$$

This quantity is unchanged in rotations

$$\mathbf{u} = [\omega \mathbf{r}], \quad v_{ik} = -\epsilon_{ikl} \omega_l, \quad u_{ik} = 0,$$

if the field \mathbf{H} is rotated simultaneously:

$$\delta \mathbf{H} = [\omega \mathbf{H}],$$

that is,

$$\epsilon_{ikl} \omega_l \frac{\partial \tilde{F}}{\partial v_{ik}} + \frac{1}{4\pi} \epsilon_{ikl} B_l \omega_k H_l = 0.$$

Hence it is easily found that

$$(\partial \tilde{F} / \partial v_{ik})_{\mathbf{H}, u_{ik}} = -\frac{1}{8\pi} (H_l B_k - H_k B_l). \quad (\text{A.1})$$

In order to obtain an expression for the stress tensor σ_{ik} , we calculate the change of the free energy, $\delta \tilde{\mathcal{F}}$, in an infinitely small displacement of the points of the body, $\delta \mathbf{u}(\mathbf{x})$. This quantity with sign reversed is equal to the work of internal forces

$$\delta R = \int f_i \delta u_i d^3 \mathbf{x}, \quad f_i = \frac{\partial \sigma_{ik}}{\partial x_k}.$$

For simplicity we shall suppose that the boundaries of the body are fixed (that is, that $\delta \mathbf{u}_{\text{bound}} = 0$) and that the deformation has no effect on the sources of the magnetic field, which are outside the body; that is, that $\delta \mathbf{j}(\mathbf{x}) = 0$.

The change of free energy can be expressed in the form

$$\delta \tilde{\mathcal{F}} = \int_{V_0} \tilde{F}'(\mathbf{x}) d^3 \mathbf{x}' - \int_{V_0} \tilde{F}(\mathbf{x}) d^3 \mathbf{x} - (4\pi)^{-1} \int_{V'} \mathbf{B} \delta \mathbf{H} d^3 \mathbf{x},$$

*It must be remembered that the probability of formation of surface nuclei is proportional to the surface of the specimen, not to its volume.

where V_0 is the volume of the body, V' is the volume outside the body, $\mathbf{x}' = \mathbf{x} + \delta\mathbf{u}(\mathbf{x})$, and $\tilde{\mathcal{F}}'(\mathbf{x}')$ is the changed value of the free energy at that point of the body which before the deformation had coordinates \mathbf{x} . In the linear approximation,

$$\delta\tilde{\mathcal{F}} = \int_{V_0} [\tilde{F}'(\mathbf{x} + \delta\mathbf{u}) - \tilde{F}(\mathbf{x}) \delta u_{ii}] d^3\mathbf{x} - (4\pi)^{-1} \int_{V'} \mathbf{B} \delta H d^3\mathbf{x}.$$

The quantity $\delta' \tilde{F} = \tilde{F}'(\mathbf{x} + \delta\mathbf{u}) - \tilde{F}(\mathbf{x})$ is the so-called material change. It is equal to

$$\delta' \tilde{F} = \frac{\partial \tilde{F}}{\partial u_{ik}} \delta u_{ik} + \frac{\partial \tilde{F}}{\partial v_{ik}} \delta v_{ik} - \frac{\mathbf{B}}{4\pi} \delta' H,$$

where

$$\delta' H_k = H'_k(\mathbf{x} + \delta\mathbf{u}) - H_k(\mathbf{x}) = \delta H_k + \delta u_i \frac{\partial H_k}{\partial x_i}$$

and use has been made of the fact that for small deformations, $\delta' u_{ik} = \delta u_{ik}$ and $\delta' v_{ik} = \delta v_{ik}$.

Thus the sum of the terms proportional to the change of magnetic field δH is

$$-(4\pi)^{-1} \int_{V_0+V'} \mathbf{B} \delta H d^3\mathbf{x} = -(4\pi)^{-1} \int \text{rot } \mathbf{A} \delta H d^3\mathbf{x} = c^{-1} \int \mathbf{A} \delta j d^3\mathbf{x} = 0.$$

The equality to zero occurs because the sources of the field have been assumed to be unchanged: $\delta j(\mathbf{x}) = 0$. Therefore $\delta\tilde{\mathcal{F}}$ can be expressed in the form

$$\delta\tilde{\mathcal{F}} = \int d^3\mathbf{x} \left[\frac{\partial \tilde{F}}{\partial u_{ik}} \frac{\partial \delta u_i}{\partial x_k} + \tilde{F} \frac{\partial \delta u_i}{\partial x_i} - \frac{1}{8\pi} (H_i B_k - H_k B_i) \frac{\partial \delta u_i}{\partial x_k} - \delta u_i \frac{B_k}{4\pi} \frac{\partial H_k}{\partial x_i} \right].$$

We note that inside the body

$$\frac{1}{4\pi} B_k \frac{\partial H_k}{\partial x_i} = \frac{1}{4\pi} B_k \frac{\partial H_i}{\partial x_k} = \frac{1}{4\pi} \frac{\partial}{\partial x_k} (H_i B_k).$$

Therefore

$$\delta\tilde{\mathcal{F}} = - \int \delta u_i \frac{\partial}{\partial x_k} \left[\frac{\partial \tilde{F}}{\partial u_{ik}} + \tilde{F} \delta_{ik} + \frac{1}{8\pi} (H_i B_k + H_k B_i) \right] d^3\mathbf{x}.$$

Hence it follows that

$$\sigma_{ik} = \frac{\partial \tilde{F}}{\partial u_{ik}} + \tilde{F} \delta_{ik} + \frac{1}{8\pi} (H_i B_k + H_k B_i). \tag{A.2}$$

Relations (A.1) and (A.2) are equivalent to relation (3.6) of the main text.

An analogous expression for the stress tensor in dielectrics was obtained (by other methods) in the book of Landau and Lifshitz.^[1]

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