Domain walls and NMR in orthoferrites

A. K. Zvezdin

(Submitted October 21, 1974)
Zh. Eksp. Teor. Fiz. **68**, 1434–1448 (April 1975)

The anisotropy of hyperfine field and of the NMR frequencies in orthoferrites are studied by symmetry theory. The shape of the nuclear magnetic resonance absorption lines is considered for nuclei located within the domain walls. The influence of a magnetic field on the line shape is studied. It is shown that the line shape depends strongly on the type of domain walls existing in the orthoferrites, so that NMR can be employed as a direct method for identifying the type of the domain wall.

PACS numbers: 76.60., 75.60.F

1. INTRODUCTION

The presence of weak ferromagnetism in orthoferrites can lead to unique features of the magnetic-resonance spectrum of nuclei located in domain walls. These features are connected with two factors: the existence in weak ferromagnets of specific types of domain walls [1-3], and the characteristic anisotropy of the dependence of the hyperfine field on the magnetic moments of the sublattices [4,5]. Orthoferrites are a convenient object for the study of this process, inasmuch as their NMR and their domain structure have been intensively investigated of late.

The magnetization in weak ferromagnets is determined by the orientation of the antiferromagnetism vector **l**. Therefore the domain wall can be characterized by the distribution of **l**(**r**). The plane in which the vector **l** rotates in the wall is determined by the character of the anisotropy. Orthoferrites are known to have domain walls of two types [2,3]. In the walls of the first type, the vector **l** rotates in the ac plane of the crystal, and the magnetization m remains essentially unchanged when **l** rotates; these walls do not differ in practice from the domain walls in ferromagnetic materials. Let the axes of the rectangular system x, y, z coincide with the axes a, b, and c of the crystal; then the vectors **l** and **m** in these walls are given by

$$\mathbf{l}=(l\cos\theta,\ 0,\ l\sin\theta),\quad \mathbf{m}=(m\sin\theta,\ 0,\ m\cos\theta),$$

where θ is the corresponding angle in the ac plane and varies in the wall from zero to π or from π to 2π. In walls of the second type, the vector **l** rotates in the ab or bc plane of the crystal, accompanied by a monotonic change of the magnetization from m to −m. In the walls of the second type we have

$$\mathbf{l}=(l\cos\varphi,\ l\sin\varphi,\ 0),\quad \mathbf{m}=(0,\ 0,\ m\cos\varphi),$$

where the angle φ changes in the ab plane from zero to π or from π to 2π. Reversal of the orientation of the vector **l** in the wall causes corresponding changes in the hyperfine field and in the NMR frequency.

2. ANISOTROPY OF HYPERFINE FIELD IN ORTHOFERRITES

To determine the NMR spectrum from the domain walls, it is necessary to know the angular dependence of the NMR frequency. It is convenient to determine in it phenomenologically, by using the symmetry of the crystal [4]. Let $\mathbf{M}_i$ (i = 1, 2, 3, 4) be the magnetization of the i-th iron sublattice, and let $\mathbf{h}_i$ be the hyperfine field acting on the nuclei of the i-th sublattice. The structure of the hyperfine fields, which is uniquely determined by the magnetic structure, remains invariant under transformations of the magnetic-symmetry group of the crystal. This means that the transformation properties of $\mathbf{h}_i$ and $\mathbf{M}_i$ coincide. It is known that the following combinations [6]

$$\mathbf{F}=\mathbf{M}_1+\mathbf{M}_2+\mathbf{M}_3+\mathbf{M}_4,\quad \mathbf{A}=\mathbf{M}_1-\mathbf{M}_2-\mathbf{M}_3+\mathbf{M}_4,$$
$$\mathbf{G}=\mathbf{M}_1-\mathbf{M}_2+\mathbf{M}_3-\mathbf{M}_4,\quad \mathbf{C}=\mathbf{M}_1+\mathbf{M}_2-\mathbf{M}_3-\mathbf{M}_4$$

transform in accordance with the one-dimensional irreducible representations of the orthoferrite group Pbnm. We introduce analogous combinations of the hyperfine fields **f**, **g**, **a**, **c**; according to the foregoing, they transform in the same manner as **F**, **G**, **A**, **C** [4].

Our task is to obtain a linear relation between $\mathbf{h}_i$ and $\mathbf{M}_i$. It is known that it can take place only between those components of $\mathbf{h}_i$ and $\mathbf{M}_i$ which transform in accordance with one and the same irreducible representation. The case of two magnetic sublattices (A = C = 0) was considered in [4]. For a more detailed consideration of the problem (in particular, of the NMR spectrum of walls of the second type with reorientation of **l** in the plane ab or bc) it is necessary to take four sublattices into account.

We use the foregoing considerations concerning the symmetry of the hyperfine fields to find the angular dependence of the NMR frequency in the case when the reorientation takes place in the planes ac and ab.

A. Reorientation of l in the Plane ac (Walls of the First Type)

Let $l_z = l\sin\theta$ and $l_x = l\cos\theta$. From the thermodynamic theory of weak ferromagnets [6,7] we have $F_x \propto C_y \propto \sin\theta$, whence

$$f_x=f_1\sin\theta,\quad g_z=g_3\sin\theta,\quad c_y=c_2\sin\theta,$$

i.e., all the quantities that transform in accordance with the irreducible representation Γ_1 are proportional to $\sin\theta$ (f_1, c_2, and g_3 are constants independent of the angle θ). Analogously, all the quantities that transform in accordance with Γ_3 are proportional to l_x, i.e., to $\cos\theta$. We have

$$f_z=f_3\cos\theta,\quad g_x=g_1\cos\theta,\quad a_y=a_2\cos\theta,$$

where f_3, g_1, and a_2 are constants independent of the angle θ. In our case, i.e., in the representation $\Gamma_1\times\Gamma_3$), only the foregoing combinations of the hyperfine fields differ from zero. Substituting them in the formula for the hyperfine fields

$$\mathbf{h}_1=\mathbf{f}+\mathbf{g}+\mathbf{a}+\mathbf{c},\quad \mathbf{h}_2=\mathbf{f}-\mathbf{g}+\mathbf{c}-\mathbf{a},$$
$$\mathbf{h}_3=\mathbf{f}+\mathbf{g}-\mathbf{a}-\mathbf{c},\quad \mathbf{h}_4=\mathbf{f}-\mathbf{g}+\mathbf{a}-\mathbf{c},\qquad (1)$$

we obtain their angular dependence. We assume also that the main contribution to the hyperfine field acting

on the i-th nucleus is made by an isotropic interaction proportional to the magnetic moment of the corresponding i-th ion (this agrees with the experimental data). This means that the structure of the hyperfine fields at the Fe nuclei differs little from the structure of the magnetic moments. The largest of the magnetic moments F, G, A, and C is G, and for the nuclei the largest is analogously g. We therefore regard ratios of the type f_1/g_1 as small parameters (of the order of 10^{-2}, as follows from the analysis of the experimental data given in [8,9]).

The NMR frequency of Fe^{57} (I = 1/2) is given by

$$\omega_i=\gamma|\mathbf{h}_i+\mathbf{H}|, \qquad (2)$$

where **H** is an external field lying in a plane perpendicular to the easy axis. We shall consider for the sake of argument the case when the easy axis coincide with the c axis of the crystal. The external magnetic field intensity is assumed to be small in comparison with the hyperfine field ($|\mathbf{H}| \ll h_i$, with $h_i \sim 5\times 10^5$ Oe in orthoferrites), and γ is the gyromagnetic ratio of Fe^{57}.

Substituting in (2) the values of the hyperfine fields (1) and confining ourselves to terms of first order in the small ratios H/h_i, f/g, etc. we obtain

$$\omega_i=\omega_0(1-\alpha\sin^2\theta-\xi_i\beta\sin 2\theta+\xi_i h\cos\theta), \qquad (3)$$

where $\xi_i = 1$ for i = 1 and 3 and $\xi_i = -1$ for i = 2 and 4; $\omega_0 = \gamma g_1$, $\alpha = (g_1 - g_3)/g_1$, $\beta = (f_1 + f_3)/g_1$, $h = H_x/g_1$.

B. Reorientation of l in the Plane ab (cb) (Walls of the Second Type)

We put $l_x = l\cos\varphi$ and $l_y = l\sin\varphi$. For the same reasons as in subsection A, we have

$$f_z=f_3\cos\varphi,\quad g_x=g_1\cos\varphi,\quad a_y=a_2\cos\varphi,$$
$$g_y=g_2\sin\varphi,\quad a_x=a_1\sin\varphi,\quad c_z=c_3\sin\varphi.$$

Substituting these relations in (1), we obtain the angular dependence of the hyperfine fields. Substituting the values of h_i in (2) and confining ourselves to the linear approximation in the ratios f_3/g_1, a_1/g_2, etc. and $H/|h_i|$, we obtain

$$\omega_i=\omega_0(1+\alpha_1\sin^2\varphi+\eta_i\beta_1\sin 2\varphi+\xi_i h_x\cos\varphi+\xi_i h_y\sin\varphi), \qquad (4)$$

where

$$\alpha=\frac{(g_2-g_1)}{g_1},\quad \beta_1=\frac{a_1+a_2}{2g_1},\quad h_x=\frac{H_x}{g_1},\quad h_y=\frac{H_y}{\sigma},$$
$$\eta_1=\eta_2=-\eta_3=-\eta_4=1,\quad \xi_1=\xi_3=-\xi_2=-\xi_4=1.$$

In the expression for the frequencies (3) and (4), it is necessary to add also the anisotropic terms due to the decrease of the magnetic moments of the sublattices in the central part of the wall as a result of temperature excitation of the Winter magnons (see, e.g., [10]). It is obvious that, just as in the case of ferromagnets, they are described in the lowest approximation by terms of the type $f(T)\sin^2\theta$, so that they can be taken into account in a phenomenological analysis by assuming the corresponding temperature dependences of the coefficients α and α_1 in formulas (3) and (4).

We note that expressions (3) and (4) for the NMR frequencies differ significantly from each other in their dependence on the magnetic field. In case A, the double degeneracy in the frequencies is reserved in the magnetic field ($\omega_1 = \omega_3$, $\omega_2 = \omega_4$), and the y component of the external fields (which is weak enough to leave the magnetic structure unchanged), has practically no effect on the NMR frequency. In case B, at $h_x \neq 0$ and $h_y \neq 0$), a complete splitting of the NMR spectrum into four lines takes place.

3. MODEL OF THE MAIN STRUCTURE AND SHAPE OF THE NMR LINE FROM THE DOMAIN WALLS

We assume that the sample is a plate whose plane is perpendicular to the easy axis of the crystal (to be specific, the c axis). The domain-structure model is in this case a system of stripe domains (stripe structure), oriented in such a way that the vector **l** in the wall rotates in definite crystallographic planes (for example, in the plane ac in case A or in the plane ab in case B). We choose the normal to the walls to be the y axis (a more general case will be considered in Sec. 6). The period of the domain structure, i.e., the distance between the centers of the domains with equal magnetization directions will be designated by D. The question of the NMR absorption line shape of crystals with a domain structure was investigated in detail in a number of papers (see, e.g., [11,12]). We present an expression for the NMR line shape, following [12], with allowance for the specifics of the NMR spectrum and the domain structure of orthoferrites.

The absorption of a radio frequency field with polarization α (α = x, y, z) is determined by the imaginary part of the NMR susceptibility $\chi''_{\alpha\alpha}$, which takes the form[1)]

$$\chi_{\alpha\alpha}''(\omega)=\sum_l \eta_\alpha^2(y_l)\chi_n''(y_l), \qquad (5)$$

where $\eta_\alpha(y_l)$ is the local gain of the field with α polarization, located at the point y_l; $\chi_n(y_l)$ is the corresponding local NMR susceptibility relative to a circularly-polarized field

$$\chi_n''=(2\chi_{n0}/N)\,\omega_n\pi\delta(\omega-\omega_n),$$

where χ_{n0}/N is the static nuclear susceptibility (per nucleus). We have assumed that the natural NMR line width is much smaller than the "inhomogeneous broadening" due to the domain wall $\Gamma_n \ll \beta\omega_0$, a situation usually realized in orthoferrites (for example, according to [13], $\Gamma_n \sim$ 10–20 kHz in $ErFeO_3$ as against an "inhomogeneous broadening" $2\beta\omega_0 \sim 1$ MHz). For the nuclei in the domain wall, the greatest importance attaches to the local gain η_z (in the considered geometry), which can be determined in terms of the susceptibility of the displacement of the domain walls (for the sake of argument we refer to walls of the first type, in which the direction of the vector **l** is given by the angle θ; the direction of the normal to the wall coincides with the y axis):

$$\eta_z\cong\frac{\omega_0}{\gamma}\frac{d\theta}{dH}=\frac{\omega_0}{\gamma}\frac{d\theta}{dy}\frac{dy}{dH}=\frac{\omega_0}{\gamma}\frac{D\chi_{dis}}{2m_s}\frac{d\theta}{dy}.$$

We have used here the known relation $\chi_{dis} = (2m_s/D)dy/dH$, where m_s is the magnetization of the material. The function θ_y in a 180° wall is determined by the character of the free energy. In this case (taking into account the magnetic field in the ab plane) it can be defined in the following manner:

$$\frac{d\theta}{dy}=-\frac{1}{\delta}f(\theta),\quad f(\theta)=\left\{\frac{1}{K_1}[F(\theta)-F(\theta_0)]\right\}^{1/2},\quad \delta=\left(\frac{A}{K_1}\right)^{1/2},$$

K_1 is the second-order anisotropy constant, $F(\theta)$ is the density of the free energy of the crystal, and θ_0 is the value of the angle θ in the domain (as $y \to \pm\infty$). In a weak magnetic field ($H \ll H_A$, where H_A is the anisotropy field) and far from the reorientation region, $f(\theta)$ takes the simple form $f(\theta) = \sin\theta$.

In the analysis of the NMR line shape one must bear in mind one more singularity of the domain walls. There exist walls (or sections of walls) in which the angle θ changes from zero to π; they have a magnetic moment directed along the x axis ($m_x = m_s \sin\theta$). There are also walls (or their sections) in which the angle θ varies from π to 2π; their magnetic moment is directed opposite to the x axis. In the absence of a field (H = 0) the energies of both types of wall are the same, a fact we shall call for brevity "degeneracy in the wall polarization." A similar degeneracy exists also in walls of the second type. In a magnetic field lying in the ab plane, the picture changes. In walls of the first type the degeneracy in the polarization is lifted the walls (or their sections) with magnetization directed against the field have a tendency to turn into walls magnetized along the field; in a sufficiently strong field it can be assumed that all the walls of the first type are magnetized along the field, i.e., we have in them $0 \leq \theta \leq \pi$.[2)]

The walls of the second type are insensitive to the field, since the magnetization in them is always perpendicular to **H**. Consequently, walls of this type retain degeneracy in the polarization in a magnetic field.

The summation over the nuclei in (5) presupposes three types of summation: over the wall polarization p, over the nonequivalent positions, and over the nuclei inside the walls, which reduces to integration with respect to the angle:

$$\sum_i \ldots = \sum_{p=1}^{2} \sum_{i=1}^{4} \frac{N}{2D} \int dx \ldots = \frac{\delta N}{2D} \sum_{p=1}^{2} \sum_{i=1}^{4} \int_0^{2\pi} d\theta f(\theta) \ldots \qquad (6)$$

Summation over the polarization is conveniently carried out in the following manner: We assume that at H = 0 the areas of the walls of both polarizations are the same; then an analogous spectrum from walls where $\theta(\varphi)$ varies from π to 2π is added to the NMR absorption spectrum from the walls where the angle θ (or φ) changes from 0 to π. This means that the summation over the polarization can be replaced by extending the limits of integration with respect to the angle in (6) from 0 to 2π. This conclusion remains in force also for walls of the second type at H = 0, since they retain the degeneracy in the polarization also in a magnetic field. For walls of the first type at $H \neq 0$ ($H \gtrsim 100$ Oe) we assume that there remains only one energywise favored wall polarization, so that the integration with respect to θ are zero and π.

After summing over the polarizations by the described method, the summation of the nonequivalent position is very simple. It turns out that the contributions made to $\chi''(\omega)$ from individual positions, integrated over the angle, are the same. We shall demonstrate this with walls of the first type as an example (H = 0). It is easily seen from (2) that the frequency of a nucleus belonging to the first or third sublattices ($\xi = 1$) and situated in that place of the domain wall where $\theta = \theta_0$, is equal to the frequency belonging to the second or fourth sublattices ($\xi = -1$) when located where $\theta = \pi - \theta_0$. It is obvious that when θ changes from zero to π the frequencies of the first and third sublattices run through the same values of the frequencies of the second and fourth sublattices, but in a different sequence. Consequently, upon integration with respect to θ, all the sublattices give the same contribution to χ''. This reasoning is valid for walls of the second type with allowance for the fact that φ ranges from 0 to 2π.

Taking all the foregoing into account and integrating with respect to angle with the aid of a δ-function, we obtain

$$\chi_{zz}''(\omega) = AP(\omega), \qquad (7)$$

where the "amplitude" of the absorption is determined by

$$A = 4\chi_{zz0}\eta_0^2 \frac{\delta}{D} \pi\omega_0 P_0, \quad P_0 = \int_0^{\pi} d\theta |f(\theta)|. \qquad (8)$$

The line shape function $P(\omega)$ normalized[3)] to unity ($\int P(\omega)d\omega = 1$) takes the following form:

1) walls of the first type:

$$P(\omega) = \frac{2}{P_0} \sum_j \frac{f(\theta_j)}{|d\omega_1/d\theta|_{\theta=\theta_j}}; \qquad (9)$$

here

$$\omega_1(\theta) = \omega_0(-\alpha \sin^2\theta - \beta \sin 2\theta + h\cos\theta), \qquad (10)$$

θ_j is one of the roots of the equation $\omega - \omega_1(\theta) = 0$, contained in the interval $0 \leq \theta < \pi$;[4)]

2) walls of the second type

$$P(\omega) = \frac{1}{2P_0} \sum_j \frac{f(\varphi_j)}{|d\omega_2/d\varphi|_{\varphi=\varphi_j}}, \qquad (11)$$

where

$$\omega_2(\varphi) = \omega_0(1 + \alpha_1 \sin^2\varphi + \beta_1 \sin 2\varphi + h_x \cos\varphi + h_y \sin\varphi), \qquad (12)$$

φ_j ($0 \leq \varphi_j < 2\pi$) are roots of the equation $\omega - \omega_2(\varphi) = 0$.

Quantities of the type $|d\omega/d\theta|$ in (9) and (11) are functions of ω and are inversely proportional to the spin density in walls having a resonant frequency equal to ω.

4. NMR SPECTRUM AT H = 0

At H = 0 the angular dependences of the NMR frequencies for walls of the first and second types, $\omega_1(\theta)$ and $\omega_2(\varphi)$, are the same; consequently, the line-shape functions $P(\omega)$ are also the same. We consider below, for the sake of argument, the case of walls of the first type.

We consider first two particular cases in which the line shape can be represented in analytic form: $\beta \gg \alpha$ and $\beta \ll \alpha$. The first case, according to experimental data [13], is realized in certain orthoferrites near the reorientation temperature, whereas the second is typical of high temperatures.

1. The case $\alpha = 0$. Here, as follows from (10), we obtain

$$\sin 2\theta = \frac{\omega - \omega_0}{\omega_0\beta}, \quad \left|\frac{d\omega_1}{d\theta}\right| = 2\omega_0\beta\cos 2\theta,$$

$$f(\theta) = \sin\theta \left[1 + \frac{K_2}{K_1}\sin^2\theta\right]^{1/2},$$

where K_2 is the second anisotropy constant and must be taken into account near the reorientation region. Substituting these values in (9) and taking into account the ambiguity of (10) [the presence of two roots in the interval $(0, \pi)$], we obtain the absorption line shape in parametric form (u is the parameter of the function):

$$P(\omega) = \frac{1}{2P_0\omega_0\beta|\cos 2u|} \cdot \left\{\sin u \left(1 + \frac{K_2}{K_1}\sin^2 u\right)^{1/2} + \cos u \left(1 + \frac{K_2}{K_1}\cos^2 u\right)^{1/2}\right\},$$

$$\sin 2u = \left|\frac{\omega - \omega_0}{\omega_0\beta}\right|, \quad 0 \leq u < \frac{\pi}{4}, \qquad (13)$$

where

$$P_0 = \begin{cases} 1 + \dfrac{1 + K_2/K_1}{(K_2/K_1)^{1/2}} \arcsin\left(\dfrac{K_2}{K_1 + K_2}\right)^{1/2}, & K_2 > 0 \\ 1 + \dfrac{1 - |K_2|/K_1}{2(|K_2|/K_1)^{1/2}} \ln\dfrac{1 + (|K_2|/K_1)^{1/2}}{1 - (|K_2|/K_1)^{1/2}}, & K_2 < 0 \end{cases} \qquad (14)$$

Formulas (13) and (14) reflect an interesting singularity of the absorption spectrum near the reorientation region: the temperature-dependent restructuring of the spectrum is possible on account of the variation of K_2/K_1 with T, i.e., on account of the change of macroscopic characteristics of the crystal (while the microscopic NMR parameters, i.e., the coefficients in (10) and (12), can remain constant). At $K_2 \ll K_1$ ($P_0 \cong 2$), formulas (13) and (14) take the simpler form

$$P(\omega)=\frac{1}{4\omega_0\beta}\frac{\sin u+\cos u}{\cos 2u},$$
$$\sin 2u=\left|\frac{\omega-\omega_0}{\omega_0\beta}\right|, \quad 0\leqslant u<\frac{\pi}{4}. \tag{15}$$

Outside the interval $\omega_0(1-\beta) \leq \omega \leq \omega_0(1+\beta)$ we have $P(\omega) = 0$. The absorption spectrum (15) is shown in Fig. 1a. This character of the absorption spectrum agrees with the experimental data of [13]. We note that the line shape is here significantly different than in the case of a ferromagnet. In particular, the absorption has here sharp maxima at the frequencies $\omega_{max} = \omega_0(1 \pm \beta)$, which correspond to the angles $\theta_1 = \pi/4$ and $\theta_2 = 3\pi/4$ in the domain wall.

2. The case $\beta = 0$. In this case the angular dependence of NMR frequency coincides with that investigated in [11,12], and consequently the line shapes also coincide (Fig. 2a):

$$P(\omega)=\frac{1}{P_0\alpha\omega_0}\frac{[1+(K_2/K_1)\sin^2 u]^{1/2}}{\cos u},$$
$$\sin u=\left(\frac{\omega_0-\omega}{\alpha\omega_0}\right)^{1/2}, \quad 0\leqslant u<\frac{\pi}{2}.$$

At $\omega_0(1-\alpha) < \omega$ and $\omega > \omega_0$ we have $P(\omega) = 0$. The singular point of the spectrum corresponds to the angle $\theta = \pi/2$ (the center of the domain wall), and the discontinuity of the function $P(\omega)$ corresponds to the angles $\theta = 0$ and π (edges of the domain wall).

3. In the general case $\alpha \neq 0$ and $\beta \neq 0$ the expressions for the line shape are cumbersome, and we confine ourselves to the determination of the singular points of the spectrum and to an investigation of the character of the line shape near these singularities. These points (points where $P(\omega) \to \infty$) are defined by the equations

$$d\omega_1/d\theta=0, \quad \omega-\omega_1(\theta)=0.$$

The solutions of these equations are

$$\theta_1=\pi-\psi/2, \ \theta_2=\pi/2-\psi/2,$$
$$\omega_1^{max}=\omega_0\left(1-\frac{\alpha-\rho}{2}\right), \quad \omega_2^{max}=\omega_0\left(1-\frac{\alpha+\rho}{2}\right), \tag{16}$$

where

$$\rho=(\alpha^2+4\beta^2)^{1/2}, \quad \cos\psi=\alpha/\rho \quad \sin\psi=2\beta/\rho.$$

The behavior of the function $P(\omega)$ near the singular points can be explained in the following manner: near the singular point θ_1 we have

$$\frac{d\omega_1}{d\theta}\cong\left(\frac{d^2\omega_1}{d\theta^2}\right)_{\theta_1}(\theta-\theta_1). \tag{17}$$

At the same time, expanding the equation about θ_1,

$$\omega=\omega_1+\frac{1}{2}\left(\frac{d^2\omega_1}{d\theta^2}\right)_{\theta_1}(\theta-\theta_1)^2,$$

eliminating with the aid of this relation the quantity $(\theta - \theta_1)$ from (17), and substituting $d\omega_1/d\theta$ in (9), we obtain

$$P(\omega)=\begin{cases}\dfrac{[1+(K_2/K_1)\sin^2\psi/2]^{1/2}\sin\psi/2}{P_0(\rho\omega_0)^{1/2}(-\omega+\omega_1^{max})^{1/2}}, & \omega<\omega_1^{max},\\ \dfrac{[1+(K_2/K_1)\cos^2\psi/2]^{1/2}\cos\psi/2}{P_0(\rho\omega_0)^{1/2}(\omega-\omega_2^{max})^{1/2}}, & \omega>\omega_2^{max}\end{cases} \tag{18}$$

where P_0 is defined by (14).

We have put here $\alpha > 0$ for the sake of argument. At $\omega > \omega_1^{max}$ and $\omega < \omega_2^{max}$ we have $P(\omega) = 0$. The height of the peak can be estimated by putting $|\omega^{max} - \omega| = \Gamma$ in (18).

Formulas (16) show that the singular points of the spectrum correspond to certain common angles θ in the domain wall, which depend on ψ, i.e., on the ratio of the constants α and β characterizing the anisotropy of the hyperfine field. The "heights" of the absorption peaks, according to (18), are also different, depending on the ratio of these constants. A good illustration of these conclusions is the temperature dependence of the absorption spectrum in $ErFeO_3$ (see [13], Fig. 1). According to [13], the coefficient α increases with increasing temperature, and therefore the low frequency branch of the spectrum shifts towards the frequency corresponding to the center of the domain wall, and the high-frequency branch shifts towards the domain frequency.

5. NMR ABSORPTION SPECTRUM AT H ≠ 0

In a magnetic field, the NMR absorption spectra for walls of the first and second type differ significantly. This is seen already from (3) and (4). First, the absorption spectrum of walls of the first type is practically insensitive to the magnetic field directed along the b axis, and the spectrum of walls of the second type is equally sensitive both to a field directed along the a axis and to a field directed along the b axis. Let us examine in greater detail the dependence of the NMR spectrum on H for both cases.

To assess the qualitative behavior, we can set the function $f(\theta)$ in formulas (9) and (11) for $P(\omega)$ equal to $\sin\theta$. This is justified under the following two conditions: a) in a sufficiently weak magnetic field ($H \ll H_A$), when the deviation of the magnetization inside the domains from the c axis can be neglected; b) at $K_2 < K_1$, i.e., outside the reorientation region.

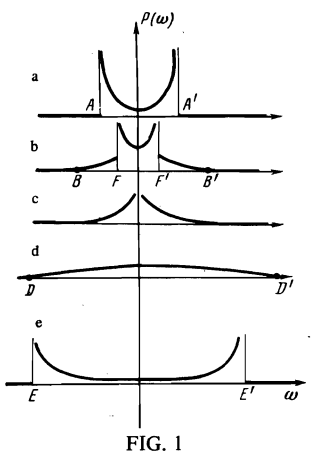

FIG. 1

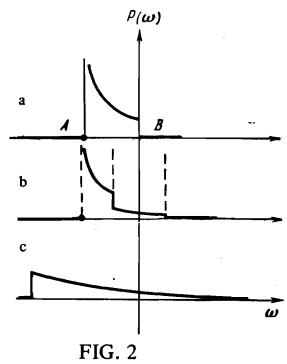

FIG. 2

FIG. 1. Schematic dependence of the line shape $P(\omega)$ and the magnetic field for walls of the first type at $\mathbf{H} \parallel$ a, $\alpha = 0$, $\beta > 0$: a) h = 0, b) $h < h_{cr}$, c) $h = h_{cr}$, d) $h > h_{cr}$, e) $h < 0$.

FIG. 2. Form of the function $P(\omega)$ at different h: a) h = 0, b) $h < h_{cr}$, c) $h > h_{cr}$. Walls of the first type, $\mathbf{h} \parallel$ a, $\beta = 0$.

A. Walls of the First Type, H Parallel to the a Axis

We consider first two limiting cases: $\alpha = 0$ and $\beta = 0$.

1. The case $\alpha = 0$. The "evolution" of the absorption spectrum[5] $P_A(\omega)$ with increasing magnetic field h is conveniently explained by regarding Eq. (10)

$$\omega/\omega_0 - 1 \equiv w = -\beta \sin 2\theta + h \cos\theta, \qquad (19)$$

as an equation of a one-parameter family of lines (in this case straight) w(h) with parameter θ. Each line of the family, corresponding to a definite value of θ, characterizes the change of the frequency of the nuclei located at that place of the wall where $\theta(y) = \theta$. It is seen from Fig. 3 that on the whole the spectrum has a tendency to spread out with increasing field h. Of greatest interest are the singular points of the spectrum, where $P(\omega) \to \infty$. The singular points of the spectrum (here—lines on the (w, h) plane) are defined by the condition $dw/d\theta = 0$. It is known that such lines are the envelopes of a family of the curves (19)—caustics. In our case the equations of the envelopes have the parametric form

$$w = -\beta(\sin 2\theta - \cos 2\theta \operatorname{ctg}\theta), \quad h = -2\beta \cos 2\theta / \sin\theta. \qquad (20)$$

The family of lines and the envelopes are shown in Fig. 3 for the case $\beta > 0$. The lines AO and OA′ ($h > 0$) correspond in (20) to the angles $\pi/4 \le \theta \le 3\pi/4$, where the lines AA correspond to the angles $0 < \theta \le \pi/4$, and the lines AE′ to the angles $3\pi/4 \le \theta < \pi$.[6] The family of lines (and the absorption spectrum) has a critical point O with coordinates $w_{cr} = 0$ and $h_{cr} = 2\beta$, to which the caustics converge. At $h > h_{cr}$, the absorption spectrum has no singular points. The conclusion that the dependence of the singular points of the domain-wall NMR absorption spectrum on the magnetic field is described by caustics of the corresponding family of frequencies, does not depend on the spectrum (19) and is general in character.

The caustic equations (20) can be represented in explicit form in the case of weak fields and near the critical field $h_{cr} = 2\beta$. We assume for the sake of argument that $\beta > 0$. The case $\beta < 0$ is obtained from the considered one by taking the coordinate transformations $w \to -w$ and $h \to -h$. The lines OAE and OA′E′ are described by the equations

$$w_{OAE} = -w_{OA'E'} = \begin{cases} -\beta + h/\sqrt{2}, & |h| \ll \beta \\ -(^8/_3)^{1/2}\beta(1-h/2\beta)^{3/2}, & h \lesssim h_{cr} \\ h, \quad h<0, & |h| \gg \beta. \end{cases}$$

At $h < h_p$ (the forms P and P′ in Fig. 3, $h_p \sim 2\beta/3$), the line sections OAE and OA′E′ coincide with the boundaries of the spectrum. At $h > h_p$, the boundaries of the spectrum (the lines PC and P′C′) are determined by the equations $w_{min} = -h$ (the line PC) and $w_{max} = h$ (line P′C′).

The character of the absorption spectrum at $P_A(\omega, h)$ (the subscripts A and subsequently B, C, and D pertain to the case under consideration) can be qualitatively established in the following manner[7]. According to (9), the function $\theta(w, h)$ (the solution of Eq. (10) determines completely the absorption spectrum. At $h < h_{cr}$, this function is not single-valued and consists of three branches that are joined together at points belonging to the envelopes; at $h > h_{cr}$, the function $\theta(w)$ is single-valued. Let us examine first how the absorption spectrum is formed at $h < h_{cr}$ (Fig. 4a). For example, at h corresponding to the line BFF′B′ in Fig. 3 we have: in the first branch θ changes from zero to $\theta_{F'}$ (the periphery of the domain wall), in the second branch we have $\theta_{F'} < \theta < \pi - \theta_{F'}$ (the center of the wall) and in the third branch $\pi - \theta_{F'} < \theta < \pi$ (periphery of the wall). The function $P_A(\omega)$, according to (9), is the sum of the contribution of all three branches, $P_1 + P_2 + P_3$ (Fig. 4b). Each of the contributions is proportional to the product of the "nuclear spin density" $|d\theta/dw|$ by the weight function $f(\theta)$. Near the envelope, the "density" $|d\theta/dw| \to \infty$, and therefore also $P(\omega) \to \infty$. From the form of the envelopes we can easily establish the variation of the function $P_A(\omega)$ with increasing field h (Figs. 1a and b). Near the envelopes, for example near the caustic OAE (Fig. 3), $P_A(\omega)$ can be represented in analytic form (by the same method as in Sec. 4):

$$P_{OAE}(\omega) = \sin\left(\frac{\pi}{4} - \frac{\sqrt{2}}{8}\frac{h}{\beta}\right)\left[2\omega_0\left(4\beta - \frac{h}{\sqrt{2}}\right)(\omega - \omega_{OAE})\right]^{-1/2}, \quad \omega > \omega_{OAE}.$$

We have put here $\theta \cong \pi/4 + h\sqrt{2}/8\beta$, which is valid when $|h| \ll \beta$. With increasing field h, the "peripheral" branches of the function $\theta(w)$ stretch out, and the branch corresponding to the center of the wall compresses and is transformed at $h = h_{cr}$ into a point (inflection on the $\theta(w)$ curve), and accordingly the function $P_A(\omega)$ has here only one singularity (see Fig. 1). The function $P_A(\omega)$ near the critical point O can be obtained in the following manner: Since $w(\theta)$ has an inflection at $\theta = \theta_{cr}$, it must be expanded in powers of $\Delta\theta = \theta - \theta_{cr}$ ($\theta_{cr} = \pi/2$) up to terms of third order, and the function $|dw_1/d\theta|$ must be expanded up to terms $(\Delta\theta)^2$. Eliminating $\Delta\theta$ from $|dw/d\theta|$ with the aid of $w(\theta)$ and substituting $|dw/d\theta|$ in (9), we obtain

$$P_A(\omega) = {}^1/_6(\omega_0\beta)^{-1/3}(\omega - \omega_0)^{-2/3}.$$

With further increase of h, the function $\theta(w)$ becomes single-valued ($|dw/d\theta| \neq 0$), the singularity at the center is transformed into a smooth maximum, and becomes gradually smeared out (Fig. 1d). With increasing field h, the function $P_A(\omega)$ decreases, and its mean value can be estimated at

$$P_A(\omega) = \frac{1}{\omega_{max} - \omega_{min}} = \frac{1}{2\omega_0 h}.$$

At the n points of the spectrum we have in this case $P_A(\omega) = 0$, since the n points correspond to $\theta = 0$, while $f(\theta) = f(\pi) = 0$.

Thus, in the considered case there are separated in the absorption spectrum two symmetrical branches of

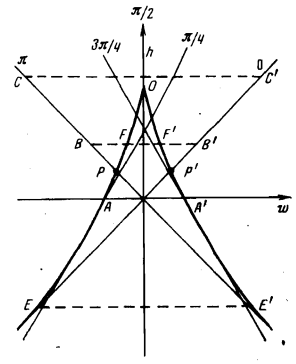

FIG. 3

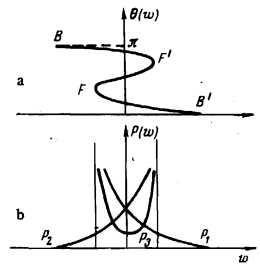

FIG. 4

FIG. 3. One-parameter family of lines $w(\theta, h)$ for walls of the first type $h \parallel a$, $\alpha = 0, \beta > 0$. Each line of this family represents the field dependence of the NMR frequency of those nuclei to which corresponds a definite angle θ in the wall. The envelopes of the family OAE and OA′E′ are the lines of the singular points of the NMR absorption spectra.

FIG. 4. Illustration of the formation of the NMR absorption spectrum: a) plot of $\theta(w)$ at $h < h_{cr}$ (see the line BFF′B′ in Fig. 3), b) contributions of different branches of the function $\theta(w)$ to the absorption spectrum $P(\omega) = P_1 + P_2 + P_3$.

singular points (Fig. 3) which converge with increasing field, and the domain-wall angles corresponding to these singular points change from $\pi/4$ and $3\pi/4$ at h = 0 to $\pi/2$ at $h = h_{cr}$. A similar dependence of the absorption spectrum on the field was observed in $ErFeO_3$ at $T = 100.3°K$[15].

2. The case $\beta = 0$. The one-parameter family of lines $w(\theta, h)$

$$\frac{\omega-\omega_0}{\omega_0} \equiv w = -\alpha \sin^2\theta + h\cos\theta \qquad (21)$$

has envelopes (singular lines of the NMR spectrum) defined by the equations (21) and $d\omega/d\theta = 0$. We put for the sake of argument $\alpha > 0$. The case $\alpha < 0$ is obtained by means of the transformation $w \to -w$ and $h \to -h$.

The envelope of the family (21) (OAO′ in Fig. 5) is determined by the equation

$$w = -\alpha[1+(h/2\alpha)^2], \quad |h/2\alpha| \leqslant 1.$$

The critical points O and O′ correspond to $h_{cr} = \pm 2\alpha$ and $w_{cr} = -2\alpha$. The function $dw/d\theta$ vanishes also on the lines $\theta = 0$ and $\theta = \pi$ (DBE and CBF in Fig. 5), but the function $f(\theta)$ in (9) also vanishes in these lines. The function $P_A(\omega)$ therefore does not become infinite on these lines but experiences a discontinuity.

Let us examine the dependence of $P_A(\omega)$ on h. At $|h| < |h_{cr}|$, the function $\theta(w)$ is not single-valued and has two branches that are joined together at points belonging to the envelope. $P_A(\omega)$ is a sum of two functions $P_1 + P_2$ corresponding to these branches (see Figs. 2a and b). Near the envelope, $P(\omega)$ tends to infinity like

$$P_A(\omega) = {}^1/_2[\alpha\omega_0(1-h^2/4\alpha^2)(\omega-\omega_{OAO'})]^{-1/2}.$$

At $h > h_{cr}$, the function $\theta(w)$ becomes single valued and $P(\omega)$, having no singularities, becomes smeared out with increasing h (Fig. 2c).

The lines EBD and CBF, where the function $P(\omega)$ experiences a discontinuity, has an interesting property that must be taken into account in the analysis of a real situation. If we introduce into the formula for the spectrum (21) a small term $-\beta\sin 2\theta$ (where $\beta > 0$ and $\beta \ll |\alpha|$), then the line FBC is transformed into a caustic, or more accurately: near the line $\theta = \pi$, there appears an envelope on which $P(\omega)$ becomes infinite like $(\Delta\omega)^{-1/2}$, while the discontinuity in the line EBD vanishes. Therefore in real situations in orthoferrites, where $\beta \neq 0$, the line FBC, in contrast to DBE, can appear as a noticeable singularity of the NMR spectrum.

3. We consider briefly the general case $\alpha \neq 0$ and $\beta \neq 0$. The singular-point lines of the NMR absorption spectrum are defined by the following equations in parametric form:

$$\frac{\omega-\omega_0}{\omega_0} \equiv w = -\frac{\alpha}{2} + \frac{\rho}{2}\cos(2\theta+\psi) - \rho\sin(2\theta+\psi)\operatorname{ctg}\theta,$$
$$h = -\rho\sin(2\theta+\psi)/\sin\theta, \qquad (22)$$

where ρ and ψ are defined by (16). The characteristic form of the envelopes of the family of the lines (22) is shown in Fig. 6.

The decisive singularity of the NMR spectrum is in the general case the presence of the critical point O. The location of the critical point on the wh plane determines the dependence of the singular point of the spectrum on the magnetic field—approach or divergence of the lines, bending of the spectral lines, etc. The coordinates of the critical point are determined by simultaneously solving (22) and the equation $d^2w/d\theta^2 = 0$, i.e.,

$$-2\beta\cos(2\theta+\psi) - h\cos\theta = 0.$$

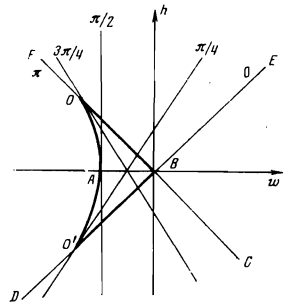

FIG. 5

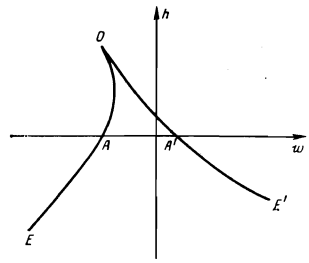

FIG. 6

FIG. 5. The family of lines $w(\theta, h)$ for walls of the first type, $\mathbf{h} \parallel \mathbf{a}$, at $\alpha > 0$ and $\beta = 0$. The envelope OO′ is the line of the singular points of the absorption spectrum.

FIG. 6. Character of the lines of the singular points of the absorption spectrum $P(\omega)$ in the general case (walls of the first type, $\mathbf{h} \parallel \mathbf{a}$, $0 < \psi < \pi/2$).

At $\alpha > 0$, $\beta > 0$ ($0 < \psi < \pi/2$) the critical point is located at $h > 0$ and is shifted towards the low-frequency end of the spectrum. At $\alpha < 0$, $\beta > 0$ ($\pi/2 < \psi < \pi$) the critical point lies in the half-plane $h > 0$ and is shifted towards the high-frequency end of the spectrum. The remaining two cases, $\pi < \psi < 3\pi/2$ and $3\pi/2 < \psi < 2\pi$ are obtained from the considered cases by the coordinate transformation $w \to -w$, $h \to -h$. Examination of Figs. 3, 5, and 6 enables us to trace the transition of the critical point from the line $\theta = \pi$ (at $\psi = 0$, Fig. 5) into the intermediate position (Fig. 6) to the line $\theta = \pi/2$ (at $\psi = \pi/2$, Fig. 3).

We present the analytic form of the plots of the singular points and of the behavior of $P_A(\omega)$ (Fig. 6) near these points (at $h/\rho \ll 1$):

$$w_{OAE} = -\frac{\alpha-\rho}{2} + h\sin\frac{\psi}{2},$$
$$P_A(\omega) = \sin\left(\frac{\psi}{2} + \frac{h}{2\rho}\cos\frac{\psi}{2}\right)\left[2\omega_0\left(2\rho - h\sin\frac{\psi}{2}\right)(\omega-\omega_{OAE})\right]^{-1/2}$$
$$w_{OA'E'} = -\frac{\alpha+\rho}{2} - h\cos\frac{\psi}{2},$$
$$P_A(\omega) = \cos\left(\frac{\psi}{2} + \frac{h}{2\rho}\sin\frac{\psi}{2}\right)\left[2\omega_0\left(2\rho - h\cos\frac{\psi}{2}\right)(\omega_{OA'E'}-\omega)\right]^{-1/2}. \qquad (23)$$

Thus, in the general case the spectrum of the walls of the first type with **H** ∥ **a** has two branches of singular points that converge to the critical point. The rate of variation of the singular points with increasing field, the position of the critical point, and the values of the absorption peaks all depend strongly on the ratio of the constants α and β which characterize the anisotropy of the hyperfine field (see (23)). An example of the considered case is the NMR spectrum in $ErFeO_3$ at $T = 128.8°K$, which was investigated in[13].

B. Walls of the First Type, h Parallel to the b Axis

In the approximation linear in $\gamma H/\omega_0$, the NMR frequencies, according to (3) are independent of H. Assuming that the shape of the walls does not depend on H, the shape of the line $P_B(\omega)$ is also independent of H. This conclusion agrees with the experimental data[7].

C. Walls of Second Type, H Parallel to the a Axis

The NMR frequency has in this case the same angular dependence as in the case A. The form of the absorption

spectrum $P_C(\omega, h)$ is determined by the superposition of two bands: P_1 from walls with $0 \le \varphi_1 < \pi$ and P_2 from walls with $\pi \le \varphi \le 2\pi$. The band P_1 coincides fully with $P_A(\omega)$ at $h > 0$, while $P_2(\omega)$ is also reduced to $P_A(\omega)$ by means of the transformation $\varphi - \pi = \varphi'$ and $h = -h'$. Thus,

$$P_C(\omega, h) = {}^1/_2[P_A(\omega, h) + P_A(\omega, -h)]. \quad (24)$$

D. Walls of Second Type, b Parallel to the b Axis

Formula (20) for the angular dependence of NMR frequency can be reduced by means of the transformation $\varphi = \pi/2 + \varphi'$ to the form ($h_x = 0$):

$$(\omega - \omega_0)/\omega_0 \equiv w = \alpha - \alpha \sin^2 \varphi' - \beta \sin 2\varphi' + h \cos \varphi'. \quad (25)$$

We see therefore that the transformation $w' = w - \alpha$, $h' = h$ transforms the spectrum (25) to the form investigated in subsection C. The absorption function for this case, $P_D(w, h)$, defined by expression (11) with $0 \le \varphi \le 2\pi$, remains invariant under the transformation $\varphi = \pi/2 + \varphi'$; it follows therefore that

$$P_D(\omega, h) = {}^1/_2[P_A(\omega + \alpha\omega_0, h) + P_A(\omega + \alpha\omega_0, -h)]. \quad (26)$$

From (24) and (26) it follows, in particular, that the absorption function $P(\omega)$ for walls of the second type has four branches of singular points in a magnetic field, two diverging with increasing field and two converging to the critical point. This fact was observed by Zalesskiĭ et al. [13] in $DyFeO_3$ near the reorientation temperature and agrees well with the fact that in $DyFeO_3$ the reorientation occurs in the ab plane of the crystal. Near the reorientation temperature, the anisotropy energy is $K_1 \approx 0$, so that walls of the second type are energywise most favored.

The fact that walls of the first type have two branches in the NMR spectrum in a magnetic field and are insensitive to a magnetic field directed along the b axis, while walls of the second type have four branches, makes it possible to use NMR to determine the type of the wall, the transformation of the domain structure, etc. (see [13]).

6. GENERALIZATION OF THE DOMAIN-STRUCTURE MODEL

We have considered the NMR spectrum using as an example a strip domain structure in the domain walls of which the vector rotate in definite crystallographic planes. The domain structure observed in orthoferrites is as a rule not of the strip type but "labryinth-like." For example, in crystals cut in such a way that the easy axis is normal to the surface of the plate, the domain walls parallel to the easy axis can become oriented in an arbitrary angle relative to the corresponding crystallographic planes (ac, bc). The results obtained above, which describe the NMR spectra in domain walls, can apparently be used also for such a domain structure. We present the justification for this conclusion.

In strongly anisotropic ferromagnets with small magnetization, the domain walls can be of more general type than the Bloch or Neel walls. Consider, for example, a wall whose plane is parallel to the c axis and makes an angle ψ with the ac plane. Let n be the coordinate along the normal to the wall. The character of the anisotropy is assumed to be such (rhombic symmetry) that the easiest magnetization plane is ac, and therefore the vector **l** (as well as **m**) rotates in it[8]; the angle θ (between **l** and **a** or between **m** and **c**) depends on n.

The energy of such a wall (per cm^2), in the standard notation, is given by

$$\sigma = \int \left\{ A \left(\frac{d\theta}{dn} \right)^2 + K_1 \sin^2 \theta - \frac{H' m_s}{2} \right\} dn, \quad (27)$$

where the magnetic field H′ is determined by Maxwell's equation

$$\operatorname{div} \mathbf{B} = \frac{\partial}{\partial n} (\mathbf{H}' + 4\pi \mathbf{m})_n = 0.$$

It follows therefore that H′ has only one component

$$H_n' = -4\pi m_n = -4\pi m_s \sin\theta \sin\psi.$$

Substituting this expression in (27) we obtain

$$\sigma = \int \left\{ A \left(\frac{d\theta}{dn} \right)^2 + (K_1 + 2\pi m_s^2 \sin^2 \psi) \sin^2 \theta \right\} dn. \quad (28)$$

A variational procedure leads to Eq. (6), where

$$\delta = \left[\frac{A}{K_1} \left(1 + \frac{1}{q} \sin^2 \psi \right) \right]^{1/2}, \quad \sigma = 4 \left[A K_1 \left(1 + \frac{1}{q} \sin^2 \psi \right) \right]^{1/2}.$$

In orthoferrites the quantity $q = K_1/2\pi m_S^2$ is much larger than unity ($\sim 10^2$), so that the energy σ of such a wall depends very little on its orientation, i.e., the walls can bend slightly, forming a labyrinth-like structure. The vector **l** remains everywhere in the ac plane. It is obvious that the NMR spectrum for such a structure is determined by formulas (7) and (8), where the reciprocal period 1/D must be replaced by S/V (S is the total area of the domain walls and V is the volume of the sample).

These results can be easily generalized to the case when account is taken of the influence of the constant K_2 and of an external magnetic field, and the main conclusions of this section remain in force then.

The author is grateful to A. V. Zalesskiĭ for interesting discussions which served as the impetus for this work and helped in its performance.

[1)] We shall investigate principally the singular points of the spectrum and their behavior following variation of the magnetic field and the temperature, so that in the analysis of the line shape we confine ourselves only to $\chi''(\omega)$, i.e., to the case of a vanishingly weak high-frequency field. Obviously, the character of the behavior of the singular point remains the same also in the more general case when partial saturation of the line takes place and its shape is determined by the combination χ' and χ''.

[2)] Experiment shows [13] that the field in which the domain-wall magnetization exists is of the order of 100 Oe. A probable mechanism of the magnetization of the walls are processes of displacement of the Bloch lines.

[3)] We note that P_0 is proportional to the domain-wall energy $\sigma_W = 2(AK_1)^{1/2} P_0$.

[4)] For walls of the first type at H = 0, the integral $\int_0^{2\pi}$ in (6) is a sum of two identical integrals $\int_0^{\pi}$ and $\int_0^{2\pi}$.

[5)] The subscript A of the function $P(\omega)$ denotes that we are dealing with the absorption function for walls of the first type with **H** ∥ a (case A).

[6)] In Sec. 3 we have established that only the case $h > 0$ has a physical meaning for walls of the first type (**H** ∥ a), in a sufficiently strong magnetic field (the walls are polarized along the field). The properties of the family of lines (19) at $h < 0$ will be used later on in an analysis of the absorption spectrum of walls of the second type.

[7)] From the mathematical point of view, the problem of the "evolution" of the absorption spectrum $P(\omega)$ as the magnetic field is varied is analogous to the problem of the appearance or vanishing of folds in the case of motion of a continuous medium (see, e.g. [14], pp. 86–93).

[8)] This is valid in the zeroth approximation in the small parameter $4\pi m_S/H_b$, where H_b is the anisotropy field that prevents the vector l from leaving the plane ac.

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Translated by J. G. Adashko
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