NMR of Fe^{57} nuclei and reorientation of spins in domains and domain walls of $ErFeO_3$ and $DyFeO_3$ crystals

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A steady-state technique is used to study the NMR of Fe^{57} nuclei in the domain walls of $ErFeO_3$ and $DyFeO_3$ crystals as a function of the temperature and of the external static field strength. A correlation between the NMR frequencies in the domains and the domain walls is found for $ErFeO_3$ in a temperature range that includes the spin reorientation region. A change in the type of domain walls with changing temperature is observed for the $DyFeO_3$ crystal. The singularities of $ErFeO_3$ and $DyFeO_3$ NMR spectra are discussed within the framework of the pehnmomenological approach developed by Zvezdin (this issue) for the special case of orthoferrites. It is shown that for orthoferrites and essentially new type (compared with conventional ferromagnets) of "interboundary" NMR spectrum is observed; it can be ascribed to a peculiar angular dependence of the NMR frequencies.

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1. INTRODUCTION

This paper is devoted to an investigation of the NMR spectra of FE^{57} nuclei (natural isotopic content) in the domain walls (DW) of the rare-earth orthoferrites $ErFeO_3$ and $DyFeO_3$. The crystals were grown by a hydrothermal method. The dimensions of the crystals and their shapes can be assessed from the photograph at the end of the article (see Fig. 6). To observe the NMR we used a superregenerative radio spectrometer.

NMR was first observed in $ErFeO_3$ crystals by the spin-echo method in [1]. A splitting of the NMR frequencies for the nuclei in the domains was observed in the spin-flip region. This fact was attributed to the onset of nonequivalence of the iron ions.[1,2] In our preceding studies, on the other hand, we observed a split NMR spectrum from the nuclei in the DW for the crystals $YFeO_3$[3,4] and $DyFeO_3$.[5] Attention is called to the fact that the frequency splitting in the domains and in the DW are almost equal. It will be shown in this paper that these two types of splitting can be explained from a common point of view if account is taken of the distinguishing features of the angular dependence of the frequencies features due to the magnetic symmetry of the orthoferrites. Such an analysis was carried out by Zvezdin.[6] The main results of this analysis are presented in Sec. 2.

In Sec. 3 we present the experimental results of an investigation of NMR in $ErFeO_3$. The measurements were carried out in a temperature interval that extends over the spin-flip region. The NMR could be observed both on the nuclei in the domains and in the DW, so that a correlation could be found between the corresponding NMR frequencies. Section 4 is devoted to an NMR investigation of the character of the spin rotation in the DW of the $DyFeO_3$ crystal. The treatment of the singularities of NMR spectra in $ErFeO_3$ and $DyFeO_3$ is based on a single theoretical approach which is described in Sec. 1 (we therefore deemed it advisable to have the theoretical part precede the experimental material). The article ends with a conclusion (Sec. 5), in which certain deductions of general character are made.

2. PRINCIPLES OF THE THEORY OF NMR SPECTRA FROM NUCLEI IN DW OF ORTHOFERRITES

For orthoferrites there have been theoretically considered two types of domain walls:[7,8] in certain walls (we shall call them, following [6], walls of type 1), the spin rotation occurs in the ac (xz) plane, while in the others (type II walls) it occurs in the ab (xy) plane. In the first case, the ferromagnetism vector $\mathbf{m}$ rotates together with the antiferromagnetism vector $\mathbf{l}$ through 180° in the ac plane. In the second case, $\mathbf{m}$ retains its orientation among the c(z) axis, but changes in absolute magnitude and passes through zero at the center of the wall.

Zvezdin,[6] starting by taking four sublattices into account, considered the NMR-spectrum singularities that are typical of DW of type I and II. Let us describe his main conclusions, which we shall need for the analysis of the experimental results. From the character of the magnetic symmetry of the orthoferrites it follows that the spin rotation in the ac plane (DW of type I) should lead to the following dependence of the NMR frequency and of the local field H_n on the angle θ between $\mathbf{l}$ and the a axis (or between $\mathbf{m}$ and the c axis, $\mathbf{l} = \mathbf{M}_1 + \mathbf{M}_3 - \mathbf{M}_2 - \mathbf{M}_4$, $\mathbf{m} = \mathbf{M}_1 + \mathbf{M}_2 + \mathbf{M}_3 + \mathbf{M}_4$):

$$2\pi\nu_i(\theta) = \gamma H_{n,i}(\theta) \approx 2\pi\nu_0(1 - \alpha\sin^2\theta - \xi_i\beta\sin 2\theta) + \xi_i\gamma H_x\cos\theta, \quad (1)$$

where $\xi_i = 1$ for the sublattices 1 and 3 and $\xi_i = -1$ for the sublattices 2 and 4, α and β are certain phenomenological hyperfine interaction constants, H_x is the component of the external constant field along the a axis, and γ is the nuclear gyromagnetic ratio. The mutual orientation of the magnetic moments of the iron ions (M_1, M_2, M_3, and M_4) is chosen such that at $\mathbf{m} \parallel z$ and $\mathbf{l} \parallel x$ their projections on the y axis are connected by the relation $M_{1y} = -M_{2y} = -M_{3y} = M_{4y}$. An expression of the type (1) was obtained independently by Zvezdin[9] and by Bar'yakhtar et al.,[2] where in the former case the authors start from general laws of magnetic symmetry of the orthoferrities, and in the latter they used a procedure with a calculation of the dipole-dipole interaction.

The expression for the NMR frequency and for H_n in the case of walls of type II is [6]

$$2\pi\nu_i(\varphi) = \gamma H_{n,i}(\varphi) \approx 2\pi\nu_0(1+\alpha_1 \sin^2\varphi + \eta_i\beta_1 \sin 2\varphi) + \xi_i\gamma(H_x \cos\varphi + H_y \sin\varphi), \quad (2)$$

where φ is the angle between **l** and the a axis in the ab plane; $\eta_i = 1$ for sublattices 1 and 2 and $\eta_i = -1$ for sublattices 3 and 4; $\xi_1 = \xi_3 = -\xi_2 = -\xi_4$.

An important circumstance is that when the spins are rotated in the ac plane the inequality of the local fields arises between pairs of spins that are oppositely directed relative to the x axis, and when the rotation is in the ab plane, the inequality is between pairs of spins that are directed in the same side. This difference in the character of the nonequivalence of the sublattices for walls of type I and II can be understood from simple symmetry considerations. Indeed, the change of the local symmetry along the thickness of a type-II wall can be represented in the form of the following sequence of magnetic point groups: m′m′m′(domain)−2/m (region between the domain and center of the DW)−mmm (the center of the DW).[1)] Thus, in a wall of type II there is conserved a simple twofold symmetry axis, which coincides with the c axis. This axis admits already of inequality of the fields (or magnetic moments) for sublattices 1 and 2 relative to 3 and 4. In a wall of type I, there is preserved the twofold antisymmetry axis 2′, which is parallel to the b axis, and admits of inequality of the fields for the sublattices 1 and 3 relative to 2 and 4 in the perpendicular plane ac.[2)] These singularities also readily demonstrate that the nonequivalence of the fields for walls of type II can be obtained analytically only when four magnetic sublattices are taken into account.

The appearance of two NMR branches at angles θ and φ different from zero and $\pi/2$ causes the integral NMR spectrum from the nuclei in walls of type I and II to have two singular points (two maxima of the NMR absorption) at frequencies which we shall designate by ν_{max} (the higher-frequency signal) and ν_{min} (the low-frequency signal). For walls of type I and II these points are determined respectively from the conditions

$$d\nu_i(\theta)/d\theta = 0, \quad d\nu_i(\varphi)/d\varphi = 0 \quad (3)$$

and correspond to the largest density of the distribution of the nuclear oscillators with respect to frequency.

A difference between the NMR spectra of walls I and II appears when an external constant field H is applied. For walls of type I, a field H ∥ b should not influence the NMR signal frequencies, since it is perpendicular to the plane in which the spins are located. It is meaningless to consider the orientation H ∥ c since at **m** ∥ c this field annihilates the domain walls. In the case H ∥ a, a significant influence on the character of the variation of the NMR frequencies is exerted by the polarization of the DW. Let us dwell on this question in greater detail.

All type-I DW can be arbitrarily divided into "right-hand," corresponding to rotation to **m** and **l** through 180° clockwise, and "left-hand" corresponding to 180° rotation counterclockwise. Mathematically this means that the angle θ can vary from zero to π (left-hand walls), or from π to 2π (right-hand walls). In a demagnetized crystal at H = 0 one should expect an equally probable distribution of sections of right and left hand walls over the volume of the crystal, since they are energy-wise absolutely equivalent. Such a state was called "degenerate with respect to polarization" in [6].

In a field H ∥ a, the equivalence of the right and left-hand walls is violated: it follows of necessity from experiment that even relatively weak fields[3)] or walls of type I are polarized in the direction of the external field (the degeneracy with respect to polarization is lifted, and the vector **m** is oriented in the DW 'favorably'' relative to the field).

When polarization is taken into account it turns out that at $H_x > 0$ all the walls become left-hand ($0 < \theta < \pi$), and when the field is reversed ($H_x < 0$) they become right-hand ($\pi < \theta < 2\pi$). It follows from (1) and (3) that in both cases the frequency ν_{max} is decreased and ν_{min} is increased, the shift of these frequencies being determined by the parameters α and β. Thus, for walls of type I, owing to the influence of the polarization in the field H ∥ a, two converging frequency branches should be observed. For walls of type II, fields $H_x \neq 0$ and $H_y \neq 0$ should cause a splitting of the NMR spectrum into four branches (each of the signals at the frequencies ν_{max} and ν_{min} splits into two). This feature is connected with the fact that in walls of type II it is the local field belonging to other sublattices that become nonequivalent in comparison with walls of type I. In walls of type II there are also possible right-hand and left-hand rotations of **l** in the ab plane, but the degeneracy with respect to polarization is not lifted in fields H_x and H_y, since the vector **m** remains perpendicular to H at all times. The presence of right-hand and left-hand walls of type II does not affect the character of the splitting of the frequencies ν_{max} and ν_{min} in fields H ∥ a or H ∥ b. In the general case (at an arbitrary direction of H in the ab plane), four branches appear for each of the frequencies.

An interesting conclusion of the theory is the presence of a certain critical field H_{cr} for the converging frequency branches. At $H > H_{cr}$ the NMR signal becomes smeared out. Attention will be called to this feature of the NMR spectra in subsequent sections of the article in connection with the discussion of the experimental results.

3. NMR IN DOMAINS AND DOMAIN WALLS, AND SPIN FLIP IN $BrFeO_3$

Figure 1 shows the results of measurement of the NMR frequencies for the crystal $ErFeO_3$ at various temperatures. The circles represent the frequencies of the resonance lines obtained when the radiofre-

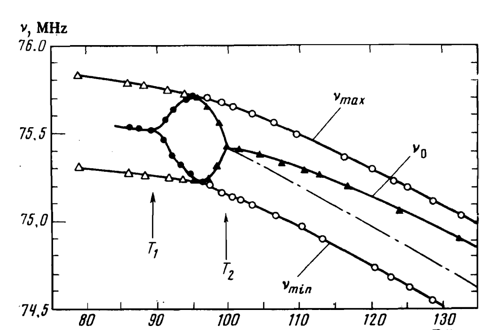


FIG. 1. Depence of NMR frequencies in domains and domain walls of $ErFeO_3$ crystal on the temperature: ○, ● – h∥c; △, ▲ – h∥a; dark points–signals from nuclei and domains, light points–signals from nuclei in domain walls.

quency (RF) field h is parallel to the rhombic axis c, while the triangles correspond to h ∥ a. At temperatures somewhat lower than T_1 (the start of the spin flip), when **m** ∥ a and **l** ∥ c, NMR signals of two types can be observed: a single NMR signal from the nuclei in the domains (dark circles) and two signals from the nuclei in the DW (light triangles). The signal from the nuclei in the domain is maximal when h ∥ c (the RF field is perpendicular to **m**). It is well observed also at relatively high values of the field h and has a width on the order of 10–20 kHz. The signals from the nuclei in the DW (the walls are parallel to the a axis at $T < T_1$) are observed at h ∥ a. The NMR lines from the nuclei in the domain walls are much wider (~50–60 kHz). Both lines are similar in shape, phase, and intensity. Their observation calls for a low level of the RF field, as is typical of NMR in domain walls. The difference between the intrawall signals and the intradomain ones becomes manifest not only in the excitation method and line width, but also in the fact that the former vanish in constant fields H < 0.5 kOe parallel to the easy axis a, while the latter are preserved in much stronger fields.

Everything said for the temperatures $T < T_1$ can be repeated for the region $T > T_2$ (T_2 is the temperature of the end of the reorientation, when **m** settles on the c axis and l ∥ a) but now in order to obtain a signal from the nuclei in the domains (dark triangles) it is necessary to have the orientation h ∥ a, and for the signals from the nuclei in the domain walls it is necessary to have h ∥ c (light circles). The splitting of the NMR frequencies for the nuclei in the domain agrees fully with the results obtained for $ErFeO_3$ by the spin-echo method.[1]

It is seen from Fig. 1 that at $T < T_1$ and $T > T_2$ the signals from the domain-wall nuclei are observed at frequencies that are continuations of the intradomain frequencies, the frequency difference between the intrawall signals corresponding exactly to the frequency difference produced in the reorientation region when the spins in the domains are rotated in the ac plane.

The data on the influence of the external field H on the frequencies of the signals from the nuclei in the domains agree with the results obtained by the spin-echo method,[2] and we therefore stop to discuss only the influence of H on the frequency of the signals from the nuclei in the domain walls. Figures 2a and 2b show the influence of a field H ∥ a on the frequencies of the NMR signals from the nuclei in the DW at temperatures 100.3° K and 128.8° K (the signal frequencies were denoted earlier by ν_{max} and ν_{min}). It is seen from Fig. 2a that near T_2 the frequencies ν_{max} and ν_{min} approach each other symmetrically. With increasing distance from T_2, the shifts of the frequencies ν_{max} and ν_{min} become asymmetrical (Fig. 2b). The frequencies corresponding to the intersection of ν_{max} and ν_{min} are noted in Figs. 1 and 2 by dash-dot lines. When the frequencies ν_{max} and ν_{min} coincide, the signal becomes indistinguishable and smeared out. The dashed lines in Fig. 2 show the frequency shift corresponding to the gyromagnetic ratio $\gamma/2\pi$ for the nuclei Fe^{57} in the case of collinearity of H and H_n. The meaning of the solid lines in Fig. 2 will become clear subsequently.

We note that a field H ∥ b has practically no influence on the frequencies ν_{max} and ν_{min}. At $T < T_1$, when **m** ∥ a, an analogous shift of the frequencies ν_{max}

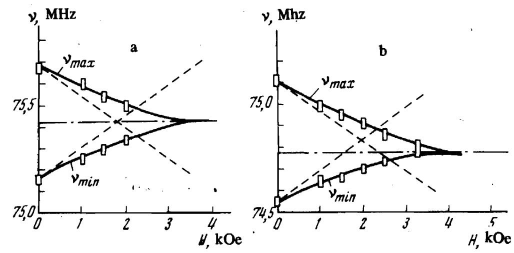


FIG. 2. Dependence of the frequencies ν_{max} and ν_{min} on the field H∥a: a–100.3°K, b–128.8°K. The solid lines were plotted in accordance with formulas (1) and (3) at the values of the parameters ν_0, α, and β indicated in the text; the dashed lines show the frequency shift corresponding to the gyromagnetic ratio $\gamma/2\pi = 0.138$ MHz/kOe for Fe^{57} nuclei.

and ν_{min} is observed, but in a field H ∥ c.

For a discussion of the results, we turn to formula (1). Inside the region from T_1 to T_2, for the nuclei in the domains, the angle θ is a function of the temperature and ranges from zero to $\pi/2$. The maximum splitting of the frequencies $2\nu_0\beta$ should be reached in this case in the midpoint of the reorientation region, when $\theta = \pi/4$, under the condition that the quantity α in (1) can be neglected (as seen from Fig. 1, this condition is satisfied). For nuclei in the domain walls, the angle θ in (1) is a function of the coordinate along the normal to the wall (walls of type I). As shown in [6], the NMR absorption spectrum has in this case singular points (maxima) at the frequencies $\nu_{max} = \nu_0(1 + \beta)$ and $\nu_{min} = \nu_0(1 - \beta)$ (see Fig. 1 in [6]), corresponding to the condition (3), i.e., at the same frequency difference $2\nu_0\beta$ as in the domains halfway in the reorientation region. It is this which explains why the difference between the intrawall signals near the reorientation region corresponds to the value of the maximum splitting of the frequencies in the domains inside the reorientation region.

The frequencies ν_{max} and ν_{min}, which correspond to the NMR absorption peaks in the domain walls, are symmetrical at $\alpha \approx 0$ relative to the frequency ν_0 corresponding to the NMR frequency in the domain and in the center of the domain wall. As seen from Fig. 1, this is confirmed by experiment for temperatures that are not too far from the reorientation region. With increasing temperature, the symmetry of the signals relative to the "domain" frequency ν_0 is disturbed (see Fig. 1): ν_{max} approaches ν_0 while ν_{min} moves away from ν_0, and the difference $\nu_{max} - \nu_{min}$ begins to increase. At the same time, the symmetry in the frequency shift in the field H ∥ a also begins to be distorted (cf. Figs. 2a and 2b). This behavior can be understood by assuming that the condition $\alpha \approx 0$ ceases to be satisfied with increasing temperature, and the NMR frequency at the center of the domain wall begins to decrease more rapidly than in the domains, owing to the increase of the α in (1). The faster decrease of the NMR frequency for the nuclei in the central part of the domain walls is predicted by spin-wave theory.[10] The NMR absorption takes in domain walls, for the general case $\alpha \neq 0$ and $\beta \neq 0$, as shown in [6], are determined as before from the condition (3), i.e., they correspond to the maximum and minimum values of the frequencies for the branches $\xi = \pm 1$ in (1). However, the violation of the condition $\alpha \approx 0$ leads, in accord with experiment to the fact that ν_{max} shifts towards frequencies corresponding to the nuclei on the periphery of the domain wall, while ν_{min}

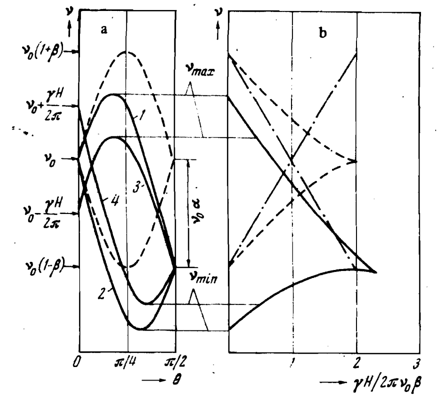


FIG. 3. Influence of a field H||a on the NMR frequencies in the domain walls: a)Depence of the frequencies for $\xi = \pm 1$ in (1) on the angle θ at $\alpha = \beta$ in the absence of a field (curves 1 and 2) and in a field H||a (curves 3 and 4). The dashed curves correspond to the case $\alpha = 0$ and H = 0. b) Corresponding shift of the frequencies ν_{max} and ν_{min} in a field H||a for the cases $\alpha = \beta$ (solid curves) and $\alpha = 0$ (dashed curves); the dash-dot line shows the shift corresponding to the gyromagnetic ratio.

shifts towards the frequency of the nuclei at the center of the domain wall. Figure 3a shows a plot of (1) for the case $\alpha = \beta$ in the angle interval $0 \leq \theta \leq \pi/2$ (curves 1 and 2), from which it is seen that ν_{max} actually shifts towards nuclei located closer to the domain (towards smaller angles θ), and ν_{min} shifts towards nuclei in the center of the domain walls. A similar conclusion with respect to localization of the nuclei responsible for the appearance of two peaks in the NMR spectrum from the nuclei in the domain walls was deduced earlier on the basis of a qualitative consideration of the NMR spectra of the crystals $YFeO_3$[4] and $DyFeO_3$,[5] but no account was taken there of the fact that the angular dependence of the NMR frequencies in the domain walls for orthoferrites differs from that for ordinary ferromagnets in the presence of a term $\pm\beta \sin 2\theta$ in (1).

To explain the character of the shift of the frequencies ν_{max} and ν_{min} in a field H || a ($T > T_2$), it is necessary to take into account the polarization of the walls (Sec. 1). The frequencies ν_{max} and ν_{min} in the presence of H || a can be determined from the condition (3) by assuming in (1) that $H_x > 0$ and $0 < \theta < \pi$ (or $H_x < 0$ and $\pi < \theta < 2\pi$). The resultant dependence of the frequencies ν_{max} and ν_{min} on H || a is shown by the solid line in Fig. 3b for the case $\alpha = \beta$. The same figure shows for comparison the shifts of the frequencies ν_{max} and ν_{min} for the case $\alpha = 0$ (dashed lines). It is seen from Fig. 3b how the increase of the parameter α influences the character of the shift of the frequencies ν_{max} and ν_{min} in a field H || a.

If we assume at $H_x \neq 0$ the simultaneous existence of the angles $0 < \theta < \pi$ and $\pi < \theta < 2\pi$ (i.e., left-hand and right-hand walls), then this leads, as can be easily verified from (1), to a splitting of each of the lines at the frequencies ν_{max} and ν_{min}. The absence of splitting is the basis for the statement that at $H_x \neq 0$ there exist only walls with polarization that is favorable with respect to H.

In fields of order 3 to 4 kOe, the NMR signals are superimposed at the frequencies ν_{max} and ν_{min}. In stronger fields, the remaining single signal is smeared out (this phenomenon was observed in [4] for $YFeO_3$).

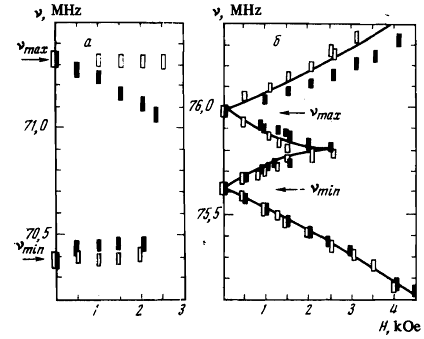


FIG. 4. Dependence of the frequencies ν_{max} and ν_{min} for $DyFeO_3$ on a field H||a (dark rectangles) or H||b (light rectangles): a—246°K, b—77°K. The solid lines were plotted in accordance with formulas (2) and (3) for the case $\alpha_1 = 0$.

The causes of the smearing of the NMR signal in fields H || a exceeding a certain critical value[4)] H_{cr} were analyzed theoretically by Zvezdin (see Figs. 1c and 1d in [6]).

By choosing the parameters ν_0, α, and β we can attempt to compare the experimentally observed shift of the NMR peaks with the theory. This is done in Fig. 2. The solid lines in this figure show the shift of the frequencies ν_{max} and ν_{min}, corresponding to the following values of the parameters in formula (1): $\nu_0 = 75.42$ MHz, $\alpha = 0$, $\nu_0\beta = 0.25$ MHz (T = 100.3 K, Fig. 2a); $\nu_0 = 74.97$ MHz, $\nu_0\alpha = 0.08$ MHz, $\nu_0\beta = 0.27$ MHz (T = 128.8 K, Fig. 2b).

4. TEMPERATURE-INDUCED RESTRUCTURING OF THE TYPE OF DOMAIN WALLS IN $DyFeO_3$

An investigation of the NMR spectra in $DyFeO_3$ crystals has shown that near room temperature the NMR spectrum from the nuclei in the domain walls has all the attributes typical of walls of type I (two branches that come close together in a field H || a, the absence of the influence of H || b). Figure 4a shows the influence of H || a and H || b on the NMR signal frequencies at 246° K. This behavior of the frequencies is typical of the case $\alpha > \beta$.

An entirely different influence is exerted on the NMR frequencies by the fields H || a and H || b at 77° K. The NMR spectrum consists as before of two lines at ν_{max} and ν_{min}, but each of them is split in the fields H || a and H || b, as shown in Fig. 4b. It was shown in Sec. 1 that the splitting in fields, $H_x \neq 0$ and $H_y \neq 0$ is a characteristic attribute of walls of type II. The solid lines in Fig. 4b show the theoretical shift of the frequencies ν_{max} and ν_{min} for the simplest case[5)] $\alpha_1 = 0$ in (2). The shift was obtained by a method similar to that used for walls of type I (Figs. 2 and 3). It is seen from Fig. 4b that if the splitting of ν_{min} is practically the same for fields H || a and H || b (this means that $\alpha_1 \approx 0$ and the signal at the frequency ν_{min} corresponds at H = 0 to nuclei for which $\varphi = n\pi/4$, n = 1, 3, 5, 7), then the splitting of the signal at the frequency ν_{max} is larger when H || b than when H || a (this means that $\alpha_1 > 0$ and the signal at the frequency ν_{max} corresponds to nuclei of the iron ions, whose spins are inclined closer to the b axis). The reasons for this disparity are not yet clear. What matters to us is that the rotation of the spins at 77° K in the ab plane, i.e., the presence of walls of type II, is subject to no doubt.

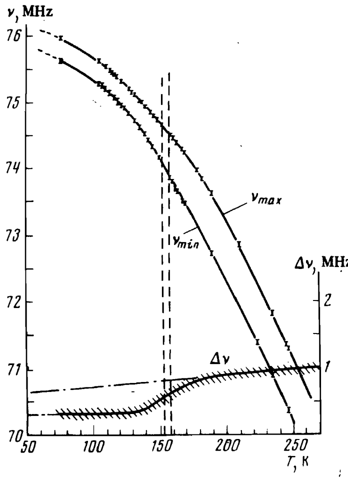
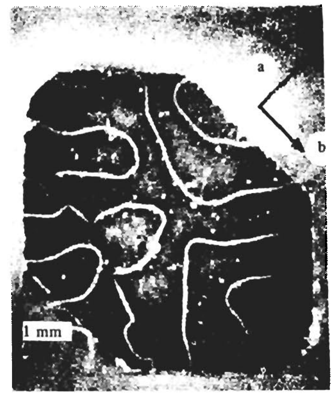


FIG. 5. Temperature dependence of the frequencies ν_{max} and ν_{min} and of the difference $\Delta\nu = \nu_{max} - \nu_{min}$ for $DyFeO_3$. The vertical dashed lines mark the temperature region where the NMR signals vanish.

FIG. 6. Domain structure of $DyFeO_3$ crystal, displayed at room temperature by the powder method, on a natural face perpendicular to the c axis. The arrows show the orientation of the rhombic axes a and b. The photograph shows how the domain walls go over to the faces inclined to the c axis.



Thus, domain walls of type I exist in $DyFeO_3$ at 246° K, and of type II at 77° K. Consequently, at intermediate temperatures, a change should take place in the character of the spin rotation in the domain walls. It was of interest to study this distinctive transition. We therefore undertook more detailed investigations of the temperature dependence of the frequencies ν_{max} and ν_{min} than in [5], and of the influence of fields H ∥ a and H ∥ b on them.

The plotting of the NMR spectra in fields H ∥ a and H ∥ b has shown that in the temperature interval 77–150° K there is observed a splitting of the frequencies ν_{max} and ν_{min}, and this is evidence that in this temperature interval there are walls of type II. At $T > 160°$ K, the splitting vanishes and the dependence of the frequencies on the fields H ∥ a and H ∥ b assumes the from shown in Fig. 4a. This means that the main process of the restructuring of the domain walls takes place in the interval 150–160° K.

The restructuring interval is particularly strongly pronounced on the temperature dependence of the frequencies ν_{max} and ν_{min}, which is shown in Fig. 5. The frequency difference $\Delta\nu = \nu_{max} - \nu_{min}$ at $T < 140°$ K (shown in the lower part of Fig. 5) is practically independent of T. In the interval 140–170° K the value of $\Delta\nu$ increases noticeably, and at $T > 170°$ K it begins to grow approximately linearly. The reason for the latter circumstance is that in the walls of type I the main contribution to the signal at the frequency ν_{max} is made by nuclei located closer to the domain, whereas the contribution to the signal at the frequency ν_{min} is made by nuclei located closer to the center of the domain wall. Spin-wave theory predicts in this case a steeper decrease of the NMR frequency with increasing T for nuclei near the center of the domain wall. The absence of a dependence of $\Delta\nu$ on T [or else a very weak dependence that lies within the limits of the measurement accuracy (the shaded region in Fig. 5)] is apparently explained for the case of walls of type II at $T < 140°$ K by the fact that the nuclei that make the main contribution to the signals at the frequencies ν_{max} and ν_{min} are approximately symmetrical with respect to the domain-wall center; they then correspond to almost the same amplitude of spin-wave excitation.

An interesting fact is that at temperatures corresponding to the narrow interval 153–158° K the NMR signals from the nuclei in the domain walls vanish almost completely. It appears that this interval corresponds to a certain intermediate (transition) state between walls of type I and II. The vanishing of the signals may be due to a strong broadening of the lines, a broadening caused by the inhomogeneous distribution of the local field at the nuclei in these intermediate walls. The temperature interval corresponds to the transition state in Fig. 5 is noted by vertical dashed lines.

Thus, the NMR data show that in $DyFeO_3$ crystals, depending on the temperature, there can exist walls of both type I and II. This peculiarity is due to the temperature-induced variation of the energy of the magnetic anisotropy. It is known that at $T = T_m \approx 40°$ K a phase transition of the Morin-point type takes place in $DyFeO_3$ and is due to the reorientation of the spins in the ab plane. If $T > T_M$ and is not too far from T_M, and ab plane remains the "easiest" from the point of view of the anisotropy energy. Naturally, the spins in the domain walls will rotate in this plane. With increasing T, the magnetic anisotropy changes in such a way that the easiest plane becomes ac (this state is typical of orthoferrites at high temperatures), making spin rotation in this plane more favored. The restructuring of the type of domain wall occurs in a relatively narrow temperature interval 150 and 160° K via a certain intermediate state (153–158° K). The NMR spectra have revealed no temperature region where walls of types I and II coexist.

In conclusion, a remark concerning the relation between the character of the spin rotation and the shape of the domain wall. Direct observation of the domain structure by the powder method has shown that the well known labyrinth-like domain structure with wavy domain walls, which is typical of all orthoferrites, is observed at room temperature in the investigated crystals (this pertains also to $ErFeO_3$ and to the previously investigated yttrium orthoferrite). Figure 6 shows an example of the domain structure for one of the investigated $DyFeO_3$ crystals. Thus, the wavy character of the domain walls does not contradict the existence of walls of type I. It can therefore be assumed that the energy requirements formulated in [8], concerning the preference for the labyrinth-like domain structure of the walls of type II, are not sufficiently stringent. This problem is analyzed in greater detail in [6].

5. CONCLUSION

The method of NMR on nuclei in DW was assumed for a long time to yield little information, owing to the great complexity of the NMR spectrum, which required averaging of a number of physical parameters over the frequency of the domain wall. A significant advance in the understanding of the intrawall NMR spectra was produced by the work of Turov and co-workers.[11] They have predicted the possibility of the appearance of two NMR signals in a uniaxial multidomain ferromagnet, one from the nuclei at the center of the domain wall and the other from the peripheral nuclei. This type of spectrum was indeed observed in a number of ferromagnets (see, e.g., [12]). We have used the conclusions of [11] in the past when a double NMR signal was observed in the crystal $YFeO_3$.[3,4] It is now clear that the reason for the appearance of two NMR peaks from the intrawall nuclei in orthoferrites is that the angular dependence of the NMR frequency is different from that of ordinary ferromagnets, although it retains certain features inherent in NMR spectra of single-lattice ferromagnets.

Thus, in the case of orthoferrites we are dealing with a fundamentally new type of NMR spectrum from the nuclei in the domain walls. A number of orthoferrite properties that favor the observation of intrawall NMR spectra (the absence of spin rotation in the domains and the possibility of preserving the domain walls in sufficiently strong fields) have made it possible to trace in detail the behavior of the resonant frequencies, to use the NMR method for unambiguous identification of the domain-wall type, and to observe the phenomenon of temperature restructuring of the spin orientation in domain walls.

A comparison of theory with experiment shows that in spite of a number of simplifying assumptions it is on the whole a correct reflection of the main features of NMR nuclei in domain walls of ferromagnets.

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[1)] In the center of a type-II wall, the magnetic structure is purely antiferromagnetic, since the noncollinearity angle vanishes.

[2)] The noncollinearity axis 2′ means its usual 2 axis in conjunction with the operation R (time reversal.

[3)] It appears that these fields are comparable in magnitude with those required to displace the DW in the case when H||C.

[4)] For the case $\alpha = 0$ we have $H_{cr} = 2\nu_0\beta 2\pi/\gamma$, see Fig. 3b.

[5)] Converging brances are also characterized here by the presence of a critical field above which the NMR signal vanishes.

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