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INVESTIGATION OF THE BOUNDARY LAYERS BETWEEN DOMAINS IN SOME FERRITES WITH THE SPINEL STRUCTURE

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Using a method based on the polar Kerr effect, an investigation was made of the width of domain boundaries, their polarity, and the magnetization distribution in them. The influence of heat treatment on the width of domain boundaries was investigated in single crystals of ferrites of the $MnFe_2O_3$ composition with an excess of manganese (25% hausmannite), of $Co_{0.94}Fe^{2+}_{0.12}Fe_{0.96}O_4$, and of $Ni_{0.54}Fe^{2+}_{0.64}Fe_2O_4$. In the iron-manganese and iron-nickel ferrites, the width of the boundaries amounted to several microns, while in the iron-cobalt ferrite it was an order of magnitude less. Graphs are given of the distribution of magnetization in the boundaries of 180° and 71° neighborhoods. It was established that in the iron-cobalt and iron-nickel ferrites, which were sensitive to heat treatment, the width of the boundaries decreased after prolonged annealing and the absolute value of the anisotropy constant increased, while the lattice parameter decreased only slightly. This effect is explained within the framework of Néel's theory.

1. INTRODUCTION

THE nature of the boundary layers between domains was investigated first by Bloch.[1] The theory was developed further by Landau and Lifshitz,[2] Néel,[3] and other workers.[4-6] Recently, a number of experimental papers have been published,[7-12] dealing with the investigation of the boundary layers between domains in metal ferromagnets.

The domain structure of ferrites can also have certain characteristic features.[13] The first investigation of the domain boundaries in ferrites was published in our previous paper.[14] The present paper is a continuation of the earlier investigation. The aim was to investigate domain boundaries in single crystals of iron-manganese, iron-cobalt, and iron-nickel ferrites and the influence of heat treatment on the changes in the boundary layers between domains.

2. SAMPLES AND MEASUREMENT METHOD

Single crystals of $MnFe_2O_4$ with an excess of manganese (25% hausmannite Mn_3O_4), of $Co_{0.94}Fe^{2+}_{0.12}Fe_{0.96}O_4$ and of $Ni_{0.54}Fe^{2+}_{0.64}Fe_2O_4$ were grown in an oxygen—hydrogen flame by the Verneuil method. These crystals were subjected to chemical and x-ray diffraction analyses to determine their composition, lattice constant, and crystallographic planes.

Spheres of 5—8 mm diameter were turned, by means of sharpened metal tubes and an abrasive suspension, from the single crystals. Two spheres were turned from each boule. One was used to investigate the influence of heat treatment on the mangetocrystalline anisotropy,[13,15] while the other was used to study the boundary layers between domains. The spheres used for the latter purpose were machined to produce the required surfaces, were carefully ground and polished, and were then boiled for 30—40 min in a 30% solution of sulfuric acid (with its concentration maintained constant during the boiling) in order to remove the surface stresses.

The boundary layers between domains were investigated by means of the polar Kerr effect. The apparatus and the method of investigation are described in detail in the published literature.[10-12] A single crystal was fixed to the stage of a microscope, on which it could be set in uniform motion. First, we used the powder method to reveal and photograph the domain structure of a single crystal and to record the location of the boundaries. Then the powder suspension was carefully removed and a modulated polarized beam of light was directed onto selected points. The sample could be moved uniformly with respect to a slit at any required velocity. When a boundary crossed the illuminated region of the sample, the field of view of the analyzer, defined by the slit, became brighter or darker (depending on the boundary polarity) and an automatic recorder recorded an appropriate signal; this effect was due to the normal component of the saturation magnetization I_{sn}.

The amplitude of the recorded curve was proportional to the magnitude of the normal component I_{sn}. The nature of the magnetization distribution in a boundary was determined from the form of the differential curve, plotted from an integral rise curve recorded on the recorder's chart when a boundary crossed the slit edge. The width of the boundary layer could be determined for any given velocity of motion of a single crystal with respect to the slit, since the slit width was 20 μ and, on the chart, represented the distance between the beginning and the end of the deviation of the curve when the boundary crossed the slit. To shorten the recording time and the length of the chart used in the recorder, the slit was set in uniform slow motion along the boundary, but was speeded up across a domain.

3. EXPERIMENTAL RESULTS

As reported before,[14] iron-manganese and iron-cobalt ferrites of the investigated composition

FIG. 1. Domain structure of a (110) plane of a single crystal of the iron–manganese ferrite.

were distinguished by the fact that they exhibited a double system of domains. It frequently happened that initially one system of domains was established, and, as the magnetic field was increased, the structure changed so that a second system appeared. The boundaries of one system intersected the boundaries of the other. A further increase in the field destroyed the initial structure; only the second system remained up to those values of the field at which the domain structure disappeared altogether. Sometimes, the double domain system could be observed in the absence of a magnetic field, if the sample was demagnetized first by commutating a magnetic field of gradually decreasing magnitude.

A structure of this type, observed on a (110) plane of the iron-manganese ferrite, is shown in Fig. 1. The white and black numbers and arrows indicate the points at which boundaries were recorded. Figure 2 shows, by way of example, the record on a chart. The numbers of the curves in Fig. 2. correspond to the white numbers in Fig. 1. Since the slit width was the same in all cases, the recorded curves indicate that the velocity of the sample was not the same. It is also evident that the boundaries had different polarities. The differential curves plotted from the integral rise curves show clearly, on a single scale, the magnetization distribution in the boundaries and the boundary polarities, and give information about the boundary widths. Such curves are given in Fig. 3a. The numbers in this figure correspond to the numbers of the curves in Fig. 2 and the white numbers in Fig. 1. It is evident from the curves in Fig. 3a that the

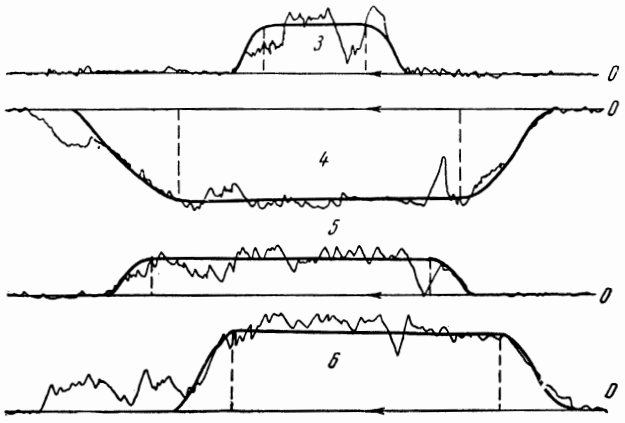

FIG. 2. Curves recorded on the recorder's chart. The numbers correspond to the white numbers shown in Fig. 1.

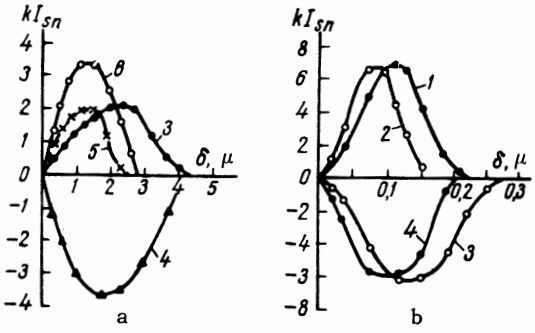

FIG. 3. Magnetization distribution in the domain boundaries of: a) iron–manganese ferrite (the numbers alongside the curves correspond to the numbers of the curves in Fig. 2); b) iron–cobalt ferrite (1 and 3 – before heat treatment, 2 and 4 – after heat treatment).

boundaries can have different widths (from 2.5 to 4.2 μ). In the principal domain structure, boundaries of different polarities and widths are observed (curves 6 and 4) with a symmetrical distribution of the magnetization in the boundary as in the case of the 180° neighborhoods.[12] Boundaries of different widths are also observed in the new structure (curves 3 and 5). The distribution of the magnetization vectors in the boundaries is asymmetric and the quantities I_{sn} have different values.

Since it is difficult to establish the same initial structure before and after annealing (the boundaries have different widths and the anisotropy constant $K_1 = -1.8 \times 10^4$ erg/cm^3, as well as the lattice constant a = 8.490 kX, change slightly due to heat treatment), it is not possible to determine the influence of heat treatment on the structure of domain boundaries in the iron-manganese ferrite. If there is any change, it must be slight.

Single crystals of the investigated iron-cobalt ferrite also exhibit double domain structure (for photographs of this structure, see[14]), but in the initial state only one system of domains is always observed. This system is retained also after heat treatment.

The magnetization distribution in the boundaries on a (110) plane of a single crystal of the iron-cobalt ferrite is shown in Fig. 3b. Curves 1 and 3 represent the domain boundaries of a sample annealed for 8 hours at 500°C. Curves 2 and 4 represent the domain boundaries of a sample annealed for 24 hours at 600°C. It is evident from these curves that the boundary width in the iron-cobalt ferrite is an order of magnitude less than in the iron-manganese ferrite. This is to be expected since the magnetocrystalline anisotropy constant of the iron-cobalt ferrite is considerably less than that of the iron-manganese ferrite. Since the boundary energy is proportional to $\sqrt{K_1}$, this energy is higher in the iron-cobalt ferrite and the boundary thickness in this ferrite is less than that in the iron-manganese ferrite.

It is also evident from these curves that the boundaries may have different polarities and different widths. The boundary width is reduced by prolonged low-temperature annealing.

Measurements of the anisotropy constants showed that, after the annealing of a single crystal of the iron-cobalt ferrite for 8 hours at 500°C, its anisotropy constant became $K_1 = 2.1 \times 10^6$ erg/cm^3, and after the annealing of the ferrite for 24 hours at 600°C, this constant reached the value $K_1 = 2.94 \times 10^6$ erg/cm^3. The lattice constant was affected only slightly by this treatment ($a_1 = 8.391$ kX, $a_2 = 8.390$ kX). Quenching from 600°C in air also increased the anisotropy constant ($K_1 = 3.5 \times 10^6$ erg/cm^3) and reduced the domain boundary width.

All these observations can be explained, at least qualitatively, by Néel's theory.[3] According to this theory, the width of a 180° boundary in a triaxial crystal is

$$\delta \approx 15\, a\sqrt{E / 6K_1}, \qquad (1)$$

and the spin orientation in the boundary is described by the equation

$$x = a\left[\frac{E}{6(K_1 + K')}\right]^{1/2} \sinh^{-1}\left(\left[\frac{K_1 + K'}{D}\right]^{1/2} \cot\vartheta\right), \qquad (2)$$

where δ is the boundary width, a is the lattice constant, E is the molecular field energy per unit volume, K_1 is the magnetocrystalline anisotropy constant, K′ and D are constants which occur in the definition of the magnetoelastic energy of a crystal. Thus, an increase in K_1 after heat treatment naturally leads to a reduction in the domain boundary width [cf. Eq. (1)].

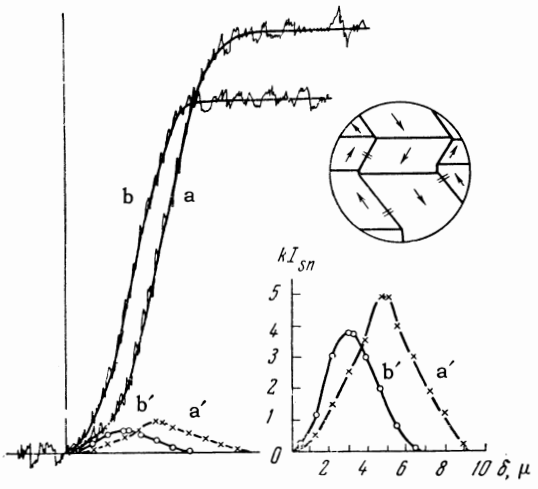

FIG. 4. Curves recorded on the recorder's chart in the investigation of the 180° boundaries in a (110) plane of a single crystal of the iron – nickel ferrite: a) before heat treatment; b) after heat treatment; a′ and b′ are the corresponding differential curves, which give the magnetization distribution in the boundaries. The schematic drawing on the right at the top of the figure shows where the boundaries were recorded.

The simplest domain structure, consisting of 180° and 71° neighborhoods, is observed in a (110) plane of a single crystal of the iron-nickel ferrite. After prolonged heat treatment (annealing at 350°C for 50 hours followed by slow cooling in the furnace in the absence of a field), the general nature of the domain structure remains the same but the boundary width decreases. Figure 4 shows curves recorded on the chart of the recorder (a—before heat treatment, b—after heat treatment). Curves a′ and b′ represent the distribution of the magnetization in 180° boundaries before and after heat treatment, respectively. The schematic diagram on the right in the upper part of Fig. 4 indicates the points at which the boundaries were recorded. It is evident from the curves in Fig. 4 that the normal component of the vector I_{sn} in the boundary is symmetrical with respect to the maximum. The boundary width is reduced by heat treatment from 9.2 to 6.7 μ.

The boundary polarity may change, which is observed on recording the polarity at different points. It is probable that the change in the boundary polarity occurs where the boundary crosses obstacles in the form of occlusions, stresses, or lattice distortions.

Figure 5 shows the magnetization distribution in 71° boundaries in the iron-nickel ferrite (a—before heat treatment and b—after heat treatment). The schematic drawing on the right indicates the points at which the boundaries were recorded. The distribution of the normal component vector I_{sn} is strongly asymmetric. Heat treatment reduces the boundary width from 3.5 to 2.4 μ.

The influence of heat treatment on the crystal lattice and the magnetocrystalline anisotropy was investigated on a single crystal of the iron-nickel ferrite.[15] X-ray diffraction analysis showed that prolonged annealing reduced the lattice parameter. Initially, the lattice parameter was $a_1 = 8.3442$ kX; after the ferrite had been annealed in air for 24 hours at 300°C and subsequently slowly cooled, the parameter became $a_2 = 8.3421$ kX; and after having annealed the ferrite for 5 days and nights, the parameter became $a_3 = 8.3415$ kX. The same value of the parameter was also obtained after five-days of annealing followed by quenching in air.

The difference between the lattice parameters of the samples annealed for 24 hours and 5 days was small, and to observe this difference, it was necessary to carry out the x-ray exposure in direct contact with the sample.

The greatest change in the anisotropy constant occurred after annealing for 2 days at 350°C. Longer annealing did not produce any marked changes. Before annealing, $K_1 = -8 \times 10^3$ erg/cm^3. After annealing for fifty hours at 350°C, we found that $K_1 = -9.9 \times 10^3$ erg/cm^3. In this case again, the annealing increased the absolute value of the first magnetocrystalline anisotropy constant.

If E is assumed to be of the order of 10^{10}, the boundary width is of the order of 10^{-4} cm and the agreement with Néel's theory is even quantitative. An increase in the anisotropy constant, and a decrease in the lattice parameter due to heat treatment, with a slight change in the molecular field per volume, leads to a reduction in the widths of the boundaries between domains.

4. CONCLUSIONS

The following conclusions may be drawn from the experiments carried out:

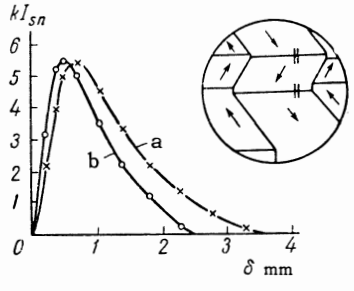

FIG. 5. Distribution of the magnetization in the 71° boundaries in a (110) plane of a single crystal of the iron – nickel ferrite (a – before heat treatment, b – after heat treatment). The schematic drawing on the right shows where the boundaries were recorded.

1. In the initial state, the boundary domain width in the iron-manganese ferrite lies within the limits 2.8—4.2 μ; in the iron-cobalt ferrite, it lies within the limits 0.25—0.35 μ; and in the iron-nickel ferrite, it lies within the limits 8.9—9.5 μ for the 180° boundaries and within the limits 3.5—4.0 μ for the 71° boundaries.

2. When there is a change in the structure of the iron-manganese ferrite of the investigated composition, a double domain system is observed. The boundaries which then appear exhibit asymmetric magnetization distribution.

3. The magnetization distribution is uniform in the 180° boundaries in the iron-cobalt and iron-nickel ferrites. In the iron-nickel ferrite, the magnetization distribution is asymmetric in the 71° boundaries.

4. The boundary polarity is of a random nature and may vary even within the same boundary that is split by obstacles.

5. In the ferrites sensitive to heat treatment (the iron-cobalt and iron-nickel ferrites), the boundary width decreases after heat treatment, the absolute values of the first magnetocrystalline anisotropy constants increase, and the lattice parameters decrease but only slightly. These changes are explained by Néel's theory.

[1] F. Bloch, Z. Physik **74**, 295 (1932).

[2] L. D. Landau and E. M. Lifshitz, Phys. Z. Sowjetunion **8**, 153 (1935).

[3] L. Néel, Cahiers de Phys. **25**, 1 (1944).

[4] S. V. Vonsovskiĭ and Ya. S. Shur, Ferromagnetizm (Ferromagnetism), Gostekhizdat, 1948.

[5] E. M. Lifshitz, JETP **15**, 97 (1945).

[6] M. Kersten, Probleme der technischen Magnetisierungskurve, Berlin, 1938.

[7] K. P. Bin, Izv. AN SSSR, ser. fiz. **21**, 1183 (1957), transl. Bull. Acad. Sci. Phys. Ser. **21**, 1174 (1957).

[8] D. J. Craik, Proc. Phys. Soc. (London) **69B**, 647 (1956).

[9] L. V. Kirenskiĭ and V. V. Veter, DAN SSSR **125**, 526 (1959), Soviet Phys. Doklady **4**, 355 (1959).

[10] V. I. Sinegubov and M. K. Savchenko, Izv. AN SSSR, ser. fiz. **25**, 599, 1449 (1961); transl. Bull. Acad. Sci. Phys. Ser. **25**, 609, 1462 (1961).

[11] M. K. Savchenko and V. I. Sinegubov, FMM **15**, 339 (1963).

[12] M. K. Savchenko and V. I. Sinegubov, JETP **44**, 781 (1963), Soviet Phys. JETP **17**, 528 (1963).

[13] Kirenskiĭ, Drokin, Dylgerov, Sudakov, and Zagirova, Izv. AN SSSR, ser. fiz. **25**, 1472 (1961), transl. Bull. Acad. Sci. Phys. Ser. **25**, 1488 (1961).

[14] Kirenskiĭ, Drokin, Dylgerov, Sudakov, and Sinegubov, Izv. AN SSSR, seriya fiz. **28**, 545 (1964), transl. Bull. Acad. Sci. Phys. Ser. **28**, 451 (1964).

[15] Drokin, Sudakov, and Gendelev, Voprosy radioélektroniki (ser. 3) **3**, 52 (1964).

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