# MAGNETIZATION PROCESSES AND DOMAIN STRUCTURE IN ORTHOFERRITE MONOCRYSTALS WITH WEAK FERROMAGNETISM

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Remagnetization properties have been studied in monocrystal orthoferrite specimens whose dimensions were close to the critical dimension for a single-domain state. It was demonstrated that, depending on the size of the magnetizing field, the determining factor in the remagnetization is: (a) in a range of fields insufficient to saturate the specimen, reversible displacement of domain boundaries; (b) in a range of fields sufficient to destroy the basic domain structure, but insufficient to remagnetize individual regions related to defects of the crystalline structure, a lag in the growth of such regions ("residual nuclei"); (c) in a range of fields producing complete saturation, a lag in the formation of remagnetization nuclei. It was shown that in cases (b) and (c), deformation of the surface layer of the crystal facilitates the remagnetization process. Subsequent annealing has an opposite effect. The regularities found in the magnetization processes in orthoferrite crystals should be observed in some degree in all ferromagnets near the critical dimension for a single-domain state.

## INTRODUCTION

DESPITE significant successes in recent years in the study of the physics of magnetization processes in ferromagnets, there has still been little investigation of the elementary events in these processes[1)]. This situation is due to the fact that in ordinary ferromagnets, the domain structure (DS) has a complicated character; it becomes considerably simplified when we go over to very small particles. In such particles, however, it is not possible to correlate individual changes in DS with the changes of magnetic properties that they produce. Yet such data are extremely necessary if we are to construct a theory of magnetization processes.

In the solution of this problem, great possibilities are opened up by the orthoferrites of the rare-earth metals and Y. In these compounds, in consequence of the slight noncollinearity of the antiferromagnetic structure, there is a spontaneous magnetic moment[1]. Because of the small magnitude of this moment and the quite large constant of natural crystallographic anisotropy, the orthoferrites have a very large critical dimension for the single-domain state (~0.5 mm). Because of this and of the magnetic uniaxiality, comparatively large crystals exhibit a simple DS, formed by only a few domains with antiparallel orientations of the magnetization[2]. In such crystals the possibility opens up of investigating magnetization processes in their details, including even such important questions as the conditions for formation and destruction of nuclei of remagnetization. It is to the study of these processes that the present paper is devoted.

## SPECIMENS AND METHOD OF MEASUREMENT

We studied monocrystal specimens of the orthoferrites $LuFeO_3$, $YFeO_3$, $YbFeO_3$, and $SmFeO_3$, obtained by the method of crystallization from the melt. The authors thank V. A. Timofeev (Institute of Crystallography, USSR Academy of Sciences) for providing the monocrystals.

The monocrystals were of nearly parallelepipedal shape and had the following dimensions: $LuFeO_3$, 3.5 × 2.2 × 3.0 mm; $YFeO_3$ (I), 1.0 × 1.0 × 3.5 mm; $YFeO_3$ (II), 1.9 × 1.5 × 2.2 mm; $YFeO_3$ (III), 1.2 × 1.0 × 1.2 mm; $YbFeO_3$, 4.0 × 4.0 × 0.8 mm; and $SmFeO_3$, 2.2 × 2.0 × 1.7 mm. The dimension along the c axis, which is the axis of easy magnetization in monocrystals of the orthoferrites $LuFeO_3$, $YFeO_3$, and $YbFeO_3$, is given last. In the orthoferrite $SmFeO_3$ the direction of easy magnetization, which at room temperature coincides with the crystallographic a axis, was oriented at an angle of about 45° to the largest face of the crystal and lay in the plane of the smallest face.

The measurements of magnetic properties in weak fields, up to 450 Oe, were made with a vibromagnetometer; in fields up to 35 kOe, by the ballistic method in an electromagnet. For magnetization in stronger fields (up to 130 kOe), use was made of apparatus for production of pulsed magnetic fields.

The DS was observed on natural faces of the crystals by the powder-pattern method: on monocrystals of the orthoferrites of Lu, Y, and Yb, on faces perpendicular to the c axis; on the monocrystal of the Sm orthoferrite, on a surface of the type (110) (the largest face of the crystal).

## RESULTS OF THE MEASUREMENTS AND DISCUSSION

1. On the surface of the monocrystals of $LuFeO_3$, $YFeO_3$, and $YbFeO_3$ in the demagnetized state, tortuous boundaries were clearly observed, separating large domains, of width from 0.3 ($YbFeO_3$) to 0.8 ($LuFeO_3$) mm, with antiparallel orientations of the magnetization. Observations on opposite faces showed that in all the crystals (including even the monocrystal of $SmFeO_3$), the DS

[1)] We shall hereafter understand by the term "magnetization processes" any changes of the magnetic state that are caused by the external field.

extended through along the direction of easy magnetization.

On application of an external magnetic field along the c axis, an appreciable displacement of the domain boundaries is observed, beginning at fields of order 1 Oe. Along with smooth movement of the boundaries, jerky movement is also observed; it is noticed that the more perfect the crystal is externally, the less pronounced the jerky motion is. The magnetization process slows down with approach to saturation. At a field equal to the saturation field, the last visually observable domain with an unfavorable orientation of the magnetization abruptly disappears. It should be mentioned that the width of the domains diminishes with diminution of the thickness of the crystals along the c axis. During magnetization, one can observe on the surface of such specimens the formation of domains of nearly cylindrical shape, as was described in reference [3]. The disappearance of such domains also occurs abruptly.

The form of the DS of the $SmFeO_3$ monocrystal is strongly influenced by the inclination of the axis of easy magnetization to the observation surface (Fig. 1). This influence shows up in a preferential orientation of domain boundaries along the projection of the a axis and an increase of the number of domains near the lower (in the figure) edge of the crystal, since the thickness of the crystal along the direction of the a axis decreases and, consequently, the density of magnetic charges increases (Fig. 1, a). This also explains the fact that on application of a magnetizing field along the a axis, the DS near this edge of the crystal is the last to disappear (Fig. 1, b-d).

The observations showed that in all the monocrystals investigated, the character of the change of DS during

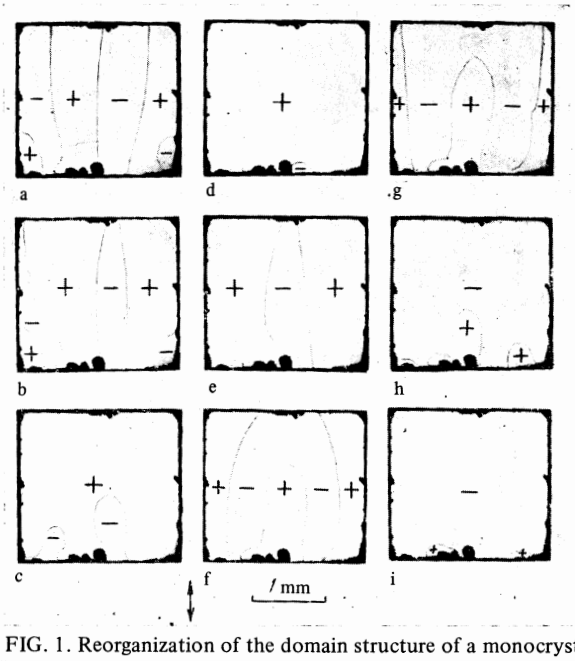


FIG. 1. Reorganization of the domain structure of a monocrystal of $SmFeO_3$ under the influence of an external magnetic field (plane of (110) type): a, H = 0; b, H = 6 Oe; c, H = 23 Oe; d, H = 30 Oe; e, H = 13 Oe; f, H = 0; g, H = −4 Oe; h, H = −23 Oe; i, H = −30 Oe. The arrow shows the direction of the projection of the a axis. The ± signs show the direction of the normal component of the magnetization in the domains.

remagnetization is determined by the maximum value of the field previously used to magnetize ($H_m$). If the field $H_m$ does not destroy the DS, then on decrease of the field, the remaining domains begin to grow (Fig. 1,e), and the remagnetization takes place by a gradual displacement of the domain boundaries (Fig. 1, f–i).

If the DS disappears completely in the field $H_m$, the remagnetization process changes qualitatively: on decrease of the field, the state of saturation persists to a certain field $H_s$, at which a domain with the opposite orientation of the magnetization abruptly originates near the edge of the crystal. With increase of $H_m$ (for $H_m > 0$), the field $H_s$ is shifted still more in the direction of negative fields, and therefore a still larger region of the crystal remagnetizes by a single jump. And when the field $H_s$ becomes larger than the saturation field, the multidomain structure becomes unstable, and the whole crystal remagnetizes by a single jump.

Such behavior of the DS was first observed on nearly single-domain monocrystalline particles of Mn-Bi alloy and was defined as a state with transitional DS [4]. Subsequently, similar phenomena were observed also on other uniaxial ferromagnets [5].

2. It is natural to expect that these peculiarities in the behavior of the DS of orthoferrites will be reflected also in their magnetic properties, including the shape of the partial cycles of hysteresis loops. Two series of such loops, measured on monocrystals $YFeO_3$ (I) and $YFeO_3$ (II), are shown in Fig. 2. (Hysteresis loops of similar shape were observed with all the investigated monocrystals.)

The measurements of hysteresis loops were made in the following order. In the initial state, the specimens were demagnetized by cooling from the Curie temperature. Then they were magnetized in field $+H_m$, and the magnetization measurement was made during a cyclic change of field: $+H_m \rightarrow -H_m \rightarrow +H_m$. Then the field $H_m$ was increased, and the next loop was measured in like manner.

The monocrystal $YFeO_3$ (I) had the most perfect faceting, and therefore the hysteresis-loop measurements on it can be most definitely correlated with the form of the DS and its reorganization in a magnetic field. The hysteresis loops measured at small values of $H_m$ (Fig. 2, A, curves a to c), not sufficient for complete disappearance of the domains in the specimen, have low values of the coercive force and of the residual magnetization. These properties of the hysteresis loops are due to the fact that, because of the perfection of the crystal structure, the domain boundaries when in motion do not encounter obstacles on their paths, and therefore the magnetization processes occur essentially by reversible boundary displacement.

Quite different in form are the hysteresis loops after magnetization of the specimen to saturation in a field $H_m = +9.4$ Oe (Fig. 2, A, curve d)[2]. In this case, the state of saturation is retained to a negative field $H_s = -5$ Oe, at which an appreciable portion of the crystal is abruptly remagnetized. The rest of the remagnetization occurs by smooth displacement of the domain boundaries all the way to field H = +4 Oe, at which the remaining domain abruptly disappears. On repeated

[2] Hysteresis loops of similar form were given in [6].

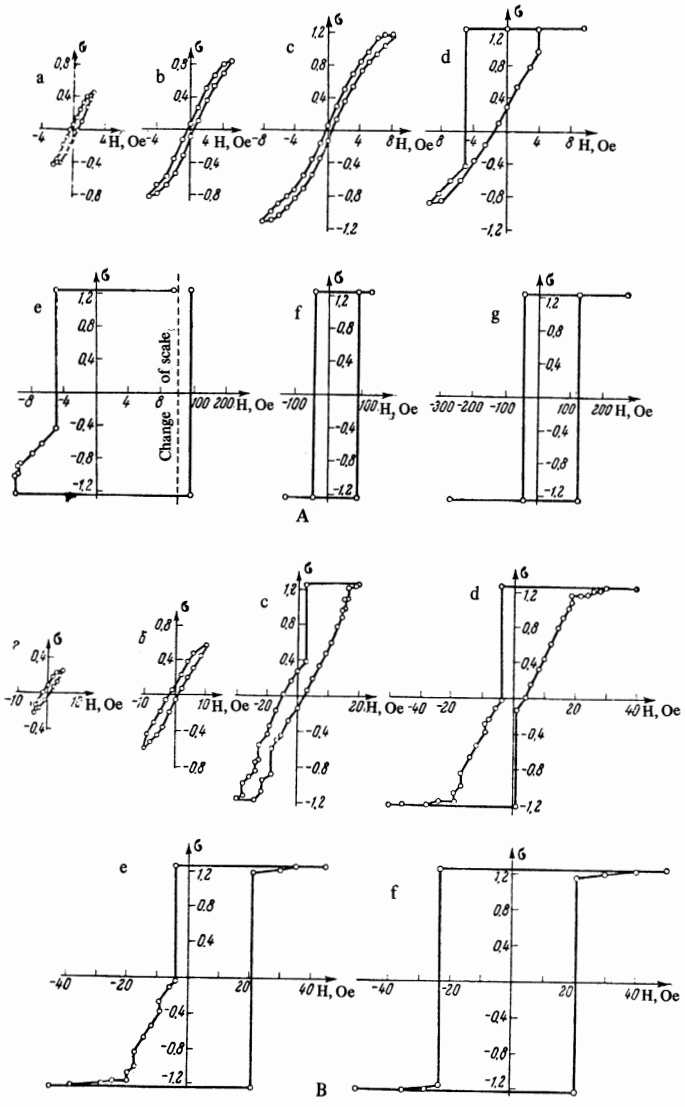


FIG. 2. Partial cycles of hysteresis loops measured on monocrystals $YFeO_3$ (I) and $YFeO_3$ (II) (A and B respectively). The values of $\sigma$ are given in G-cm$^3$/g.

remagnetization from the same field $H_m$ = +9.4 Oe, increase of the negative field to −10.2 Oe leads to a discontinuous transition to the state of negative saturation (Fig. 2, A, curve e). Remagnetization from this state occurs by a single jump at field $H_S$ = +88 Oe, which appreciably exceeds the saturation field. As a result of this, the last hysteresis loops become completely rectangular, though remaining asymmetric with respect to $H_S$ (Fig. 2, A, curves f and g). Another important fact is that with increase of $H_m$, there is observed an irregular increase of $H_S$.

In less perfect crystals (Fig. 2, B), the form of the hysteresis loops is similar in its general features to that presented in Fig. 2, A. The magnetization processes themselves, however, occur under more complicated conditions. Thus in Fig. 2, B, curves c to e, there can be observed on the branches of the hysteresis loops jumps of magnetization, caused by obstacles that the moving boundaries encounter on their paths. The incomplete remagnetization of the specimens in the case of rectangular loops (Fig. 2, B, curve f) can be attributed to the presence of hard-to-remagnetize regions, due to crystal defects of various kinds.

The observations of DS and the measurements of partial hysteresis loops showed that there is a tendency toward increase of the jump field $H_S$ with increase of the field $H_m$. It seemed of interest to elucidate the details of this phenomenon over a wider range of fields (to 130 Oe).

The function $H_S(H_m)$ for the monocrystal $YFeO_3$ (III) is shown in Fig. 3. The following fundamental characteristics of these curves should be mentioned: (a) the increase of $H_S$ with increase of $H_m$ up to a certain field $H_m^0 \approx 35$ kOe, and the presence of asymmetry of the hysteresis loops in this range of fields; (b) the steplike form of the function $H_S(H_m)$; (c) the absence of an increase of $H_S$ in fields above $H_m^0$, and the presence of symmetry of the hysteresis loops in this range.

3. The regularities stated can be explained qualitatively as follows. We assume for simplicity that in the demagnetized state there are in the specimen only two magnetic domains, with opposite orientations of the magnetization $J_S$ (Fig. 4, a). On application of a sufficiently large external magnetic field $+H_m$ ($H_m \gtrsim H_d$, where $H_d$ is the saturation field of the specimen), the specimen becomes magnetized practically to saturation. There remains unremagnetized only a very small part of the crystal, for example the part A in Fig. 4, b. We shall hereafter call such parts "residual nuclei." On diminution of the external field, because of the presence of defects, a residual nucleus remains stable to a certain field $H_{S1}$ (its starting field). At this field, the residual nucleus grows abruptly; this leads (in case $H_{S1} < H_d$) to a discontinuous remagnetization of a part of the specimen (Fig. 4, c). Subsequent change of the field to $-H_m$ leads to a practically complete remagne-

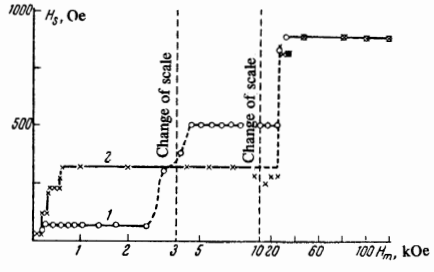


FIG. 3. Dependence of the starting field $H_S$ on the magnetizing field $H_m$, as measured on monocrystal $YFeO_3$ (III): curve 1, $+H_S(-H_m)$; curve 2, $-H_S(+H_m)$.

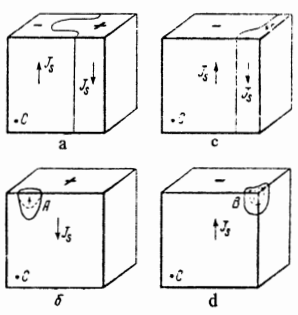


FIG. 4. Schematic drawing illustrating the formation of residual nuclei: a, H = O; b, H = $+H_m$; c, H = $H_{sl}$; d, H = $-H_m$.

tization of the specimen. In this process a new residual nucleus is formed at defects, but in still another place in the crystal (B, in Fig. 4, d). Subsequent remagnetization, of course, occurs by growth of this nucleus at its starting field $H_{S2}$, in general not equal to $H_{S1}$. When $H_S > H_d$, the inequality of the starting fields leads to asymmetry of the rectangular hysteresis loops.

The increase of $H_S$ with increase of $H_m$ and the steplike character of the functions $H_S(H_m)$ presuppose the existence for each nucleus of a number of stable states and of irreversible changes of its dimensions related to these states (shown with dashes in Fig. 4, b and d). In real crystals there may be formation not of one but of a large number of residual nuclei. Their successive destruction in the field can also be a reason for the steplike increase of $H_S$.

Thus, in the field interval under consideration, all the characteristics of the magnetization process are determined by the conditions for destruction and growth of residual nuclei, which are remnants of the original domain structure that survive near various kinds of crystal defects.

In strong fields ($H_m > H_m^0$), a nucleus of remagnetization will always originate in that place in the crystal where, because of local conditions, there is the smallest effective anisotropy field and the largest demagnetizing field (for example, at the point C in Fig. 4). Therefore the magnitude of the field $H_S$ will remain constant, independently of the magnitude and direction of the field $H_m$ (symmetry of the hysteresis loops). In this case the field $H_S$ can be identified with the field for formation of a nucleus ($H_n$), of course under the condition that the starting field of a nucleus that has originated is smaller than the field necessary for its formation.

4. To elucidate the role of the crystal surface in the formation of nuclei, measurements of the curves $H_S(H_m)$ were made (Fig. 5) on the monocrystal $YFeO_3$ (III) immediately after mechanical polishing and burnishing (curve 1) and after subsequent annealing at temperature 1000°C for two hours (curve 2). From a comparison of Fig. 3 and Fig. 5 it follows that in comparison with the original specimen, deformation of the surface layer leads to the appearance of a larger number of residual nuclei, which during remagnetization give a practically continuous series of starting fields. This causes both the smooth form of curve 1 (Fig. 5) and also the inappreciable asymmetry of the hysteresis loop. Subsequent annealing restores the crystal lattice in this

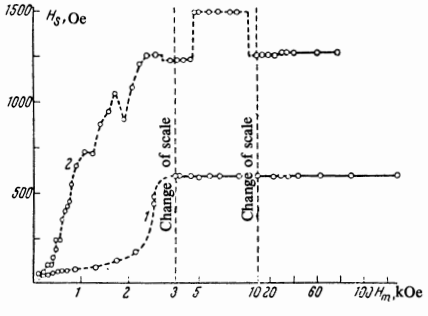


FIG. 5. Same as Fig. 3 but after polishing of the surface (curve 1) and after subsequent annealing at 1000°C (curve 2).

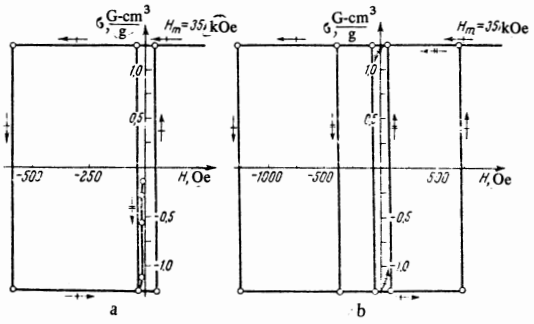


FIG. 6. Hysteresis loops obtained by cyclic demagnetization of monocrystal $YFeO_3$ (III) in the following states: a, after polishing; b, after subsequent annealing at 1000°C.

layer, which leads to a diminution of the number of potential places of formation of residual nuclei. This is indicated by the steplike form of curve 2 and by the increase of asymmetry of the hysteresis loop.

From a comparison of these same figures it is clear that the field $H_n$ also depends on the perfection of the structure of the surface layer. Thus deformation lowers $H_n$, by comparison with the original specimen, from 920 to 590 Oe. Subsequent annealing leads to an increase of $H_n$ to 1260 Oe.

On the basis of the ideas presented above about residual nuclei, one can understand also the nature of the processes that occur during cyclic remagnetization of the monocrystal $YFeO_3$ (III) (Fig. 6). Figure 6, a shows how the partial cycles change in a specimen with a deformed surface (the specimen was initially magnetized in a field +35 000 Oe). If after each abrupt remagnetization, without increasing the field, one changes it to the opposite direction, then the demagnetization process can be described as follows (H in Oe): +35 000 → −590 → +55 → −33—multidomain state. Demagnetization of the specimen after anneal proceeded by the following partial cycles (Fig. 6, b): +35 000 → −1260 → +720 → −370 → +80 → −65 → +80 → −65 etc.; that is, after attainment of a rectangular loop with $+H_S$ = 80 Oe and $-H_S$ = 65 Oe, further demagnetization of the specimen is possible only by cooling from the Curie temperature.

## BASIC RESULTS

On the basis of the experimental results presented, the following conclusions can be drawn.

On orthoferrite monocrystals of dimensions 1 to 4 mm, in which the equilibrium state corresponds to a multidomain structure consisting of a few domains (domain width ~0.5 mm), the magnetization processes were followed in detail by correlating the visually observed DS with the magnetic properties. In such crystals, with magnetization along the axis of easy magnetization, it is possible to distinguish three ranges of external field, after whose action the remagnetization processes occur successively.

a) A range of fields that do not lead to destruction of the DS. Remagnetization occurs by displacment of domain boundaries. Magnetic hysteresis is caused only by delay of the displacement processes.

b) A range of fields in which the DS disappears, but

residual domains persist. Magnetic hysteresis is caused by delay of the growth of residual nuclei and by irreversible displacement processes.

c) A range of fields after application of which magnetic hysteresis is due only to delay of the process of formation of nuclei.

The remagnetization processes depend on the perfection of the crystal structure of the specimens. In field range a), crystal defects increase the role of irreversible displacement processes; in field ranges b) and c), defects facilitate the remagnetization processes: in the first case they guarantee the possibility of formation of residual nuclei, in the second they lower the field for formation of a nucleus.

The regularities of the magnetization processes established in this work on orthoferrite crystals should occur also in other ferromagnets. Clearly, however, these regularities manifest themselves only in magnetically uniaxial ferromagnetic specimens whose dimensions are close to the critical dimension for the single-domain state.

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