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Mechanisms of magnetic and temperature hysteresis in $ErFeO_3$ and $TmFeO_3$ single crystals

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Magnetic hysteresis is studied in the orthoferrites $ErFeO_3$ and $TmFeO_3$ using the single crystal samples of millimeter dimensions. It is shown that in both materials one observes a temperature transition manifesting itself through the temperature hysteresis of the magnetic moment and a peculiar temperature evolution of the field hysteresis loop shapes near this transition. Experiments rule out the hypothesis that the ordering of the orthoferrite's rare-earth magnetic moments plays an important role in these phenomena. The hysteresis curves can be explained by a few-domain magnetic state of the samples that results from the weak ferromagnetism of the orthoferrites. The phenomenon is generic for weak ferromagnets with temperature dependent magnetization. A large characteristic magnetic length makes the behavior of the relatively big samples analogous to that observed in the nanosize samples of strong ferromagnets. © *2010 American Institute of Physics*. [doi:10.1063/1.3499616]

I. INTRODUCTION

It was reported recently[1] that the magnetic moment M of a single crystal orthoferrite $ErFeO_3$ with sample dimensions $3.9\times3.9\times3.1\ \mathrm{mm}^3$ exhibits an unusual temperature transition. A jump and hysteresis of the magnetic moment $M(T)$ are observed in a constant weak external magnetic field H. Temperature dependencies $M(T)$ for different values of H are shown in Fig. 1. A triangular temperature hysteresis is observed in the interval 5–25 K for the external field magnitudes $H=50-200$ Oe. The hysteresis disappears in higher fields—a behavior similar to that of the first order transition at the compensation point $T_c=46$ K (the latter shows up in Fig. 1 as a "butterfly pattern"[2] near T_c). It was also observed[1,3] that this behavior of $M(T)$ correlates with a qualitative changes in the hysteresis loop shapes $M(H)$ that occur as the temperature is decreased from the compensation point down to the temperatures below 25 K.

Importantly, below the compensation point of $ErFeO_3$ the decrease in temperature is accompanied by an increase in material's magnetization M_s. The triangular hysteresis $M(T)$ and the change in the $M(H)$ loop shapes were theoretically linked[3] to this magnetization growth. The explanation was as follows. Due to the weak ferromagnetism of $ErFeO_3$ the magnetic domains in the material are large. Consequently, a sample with relatively large dimensions in the millimeter range has a very simple domain structure with just a few domains. Such magnetic configurations are similar to those displayed by the nanosize samples made of strong magnetic materials like iron or cobalt. A theoretical model with a two-domain structure was quite successful at describing the experiments.[3] It had shown that the triangular temperature hysteresis corresponds to a transition from a single-domain magnetic state at higher temperatures to a few-domain state at low temperatures. In general, the idea of a few-domain state suggested that the millimeter-size samples of $ErFeO_3$ can be used to model the behavior of strong nanomagnets and possibly aid the experimental investigations of the latter.

The goal of the present paper is to show that the triangular temperature hysteresis $M(T)$, the peculiar shapes of the hysteresis loops $M(H)$, and the few-domain ground states are generic features of the materials with low, temperature dependent magnetization.

II. EXPERIMENTAL RESULTS AND DISCUSSION

The measurements were performed on the single crystals of $ErFeO_3$ and $TmFeO_3$. We used three samples of $ErFeO_3$ with different weights: a rectangular sample No. 1 with a weight of $m_1=0.4047$ g, and two samples Nos. 2 and 3 with irregular shapes and weights $m_2=0.0372$ g and m_3

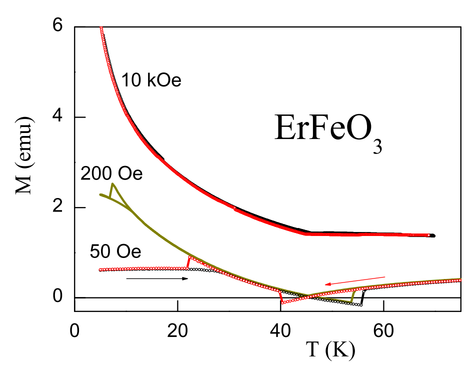

FIG. 1. (Color online) Temperature dependence of the magnetic moment projection M on the a-axis for the $ErFeO_3$ sample No. 1 at different applied magnetic fields $H\|a$. Triangular hysteresis $M(T)$ is seen below 25 K at $H=50$ Oe and $H=200$ Oe. The "butterfly pattern" between 40 and 60 K marks the first order transition at the compensation point.

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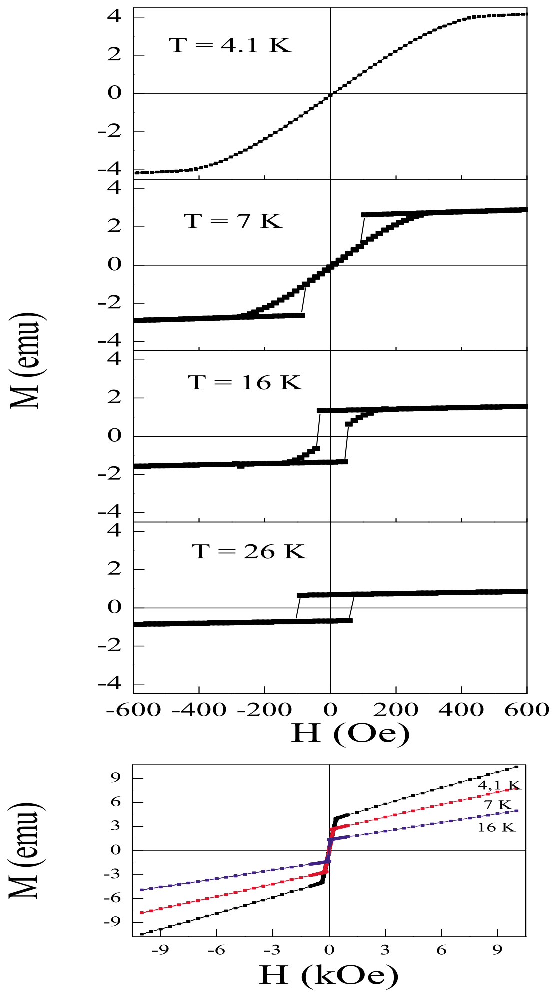

FIG. 2. (Color online) Upper panel: family of hysteresis loops for the $ErFeO_3$ sample No. 1. Four characteristic loop shapes are shown. Lower panel: general view of magnetic moment behavior in a wide field interval of ± 10 kOe.

$=0.0077$ g, respectively. One sample of $TmFeO_3$ (sample No. 4) with an oval shape and weight $m_4=0.003$ g was used. The dc value of the magnetic moment M was measured using a superconducting quantum interference device magnetometer Quantum Design MPMS-5 (temperature range 1.7–400 K, magnetic field range 0–50 kOe) and EGG Princeton Applied Research vibrating sample magnetometer (VSM) model 1200 (temperature range 4.2–280 K and field range 0–15 kOe). The magnetic field was always applied along the crystallographic a-axis that corresponds to the easy magnetization direction for the materials and temperatures studied in this work. The projection of the magnetic moment on the same axis was measured.

Figures 2–4 show the hysteresis loops for the $ErFeO_3$ samples Nos. 1, 2, and 3, respectively. The measurements were done in a wide range of temperatures below T_c. The data show that the magnetic behavior of all samples is similar. The following conclusions can be made:

(1) In the temperature interval with the lower end at approximately 20 K and upper end at T_c the hysteresis loops $M(H)$ are rectangular. The experimentally observed magnitudes of reversal fields rule out the uniform rotation mechanism (see Ref. 3 for details). The loop shapes can be explained by a magnetic reversal through the domain wall motion.
(2) As the temperature is decreased, the hysteresis loops first acquire a shape with "triangular tails," and then a shape with two separate triangles connected by a tilted central branch.
(3) Further lowering of the temperatures reduces the size of the triangular hysteresis parts of the loops, until they completely disappear. At the same time the span of the central reversible branch increases. At the end of the process, the $M(H)$ curve shows no hysteresis at all and consists of the tilted central branch and the high field branches with the residual slopes determined by the paramagnetic susceptibility of the rare-earth ions.
(4) The central branches of all hysteresis loops $M(H)$ are straight lines going through the origin with the same temperature-independent slope.

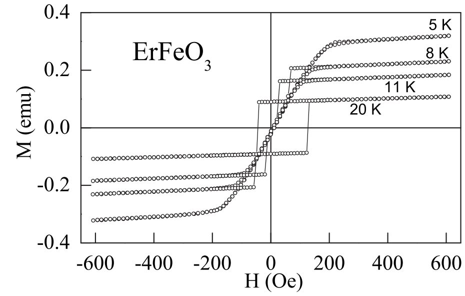

FIG. 3. Family of hysteresis loops for the $ErFeO_3$ sample No. 2.

Those experimental results are completely consistent with the conclusions of Ref. 3 and can be explained with the theoretical model suggested there. The hysteresis loops in our orthoferrite samples of relatively large sizes can be explained by the motion of just one domain wall. The central branch represents the reversible wall motion, while the jumps can be explained by the nucleation of the wall. The simplicity of the magnetic structure is the consequence of the small magnetization of an orthoferrite which results in the large magnetic domains. The shape of the hysteresis loop is determined by the relationship between the field required to push the domain wall out of the sample (expulsion field) and the field needed to nucleate it again. The ratio of the two fields is temperature dependent (see Fig. 3 from Ref. 3) and

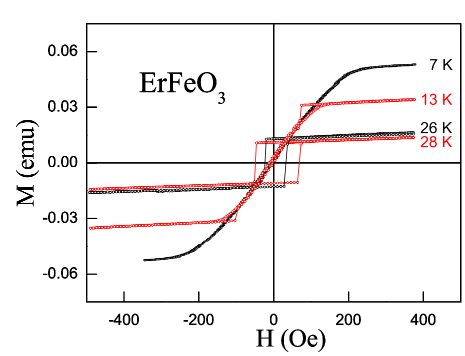

FIG. 4. (Color online) Family of hysteresis loops for the $ErFeO_3$ sample No. 3.

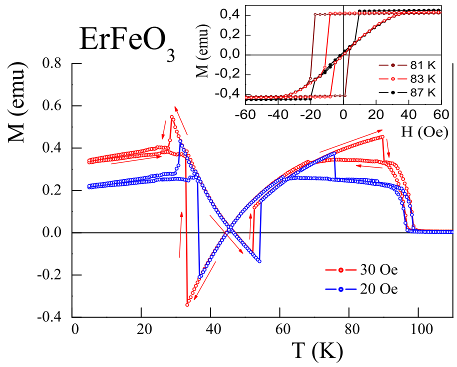

FIG. 5. (Color online) Magnetic moment of $ErFeO_3$ (sample No. 1) in a weak external field above and below T_c. Triangular hysteresis occurs at the same value of magnetization on both sides of the compensation point. Inset: $M(H)$ hysteresis loops in the interval of the triangular temperature hysteresis above T_c exhibit a triangular-tail shape similar to those found in the hysteresis interval below T_c (Fig. 2). In both cases these loop shapes determine the triangular shape of the $M(T)$ hysteresis.

governs the transformation of the loop's shapes as the temperature is changed. The exact values of the expulsion and nucleation fields, and the slope of the central branch depend on the sample shape.[3] However, our experiments show that the qualitative picture remains the same for all shapes.

The experimental results reported in this paper provide additional arguments in favor of the few-domain theory. Reference 1 discussed a possible alternative cause of the triangular temperature hysteresis at low temperatures. It was noted there that the lower end of the temperature interval 5–25 K, where this hysteresis is observed, is very close to the second Neel phase transition temperature $T_{N2}=4.1$ K where the rare-earth magnetic moments get ordered.[4–6] It was speculated that proximity to T_{N2} could somehow produce the phenomenon. Below we present experimental results which rule out this scenario.

Within the model of Ref. 3 the only cause of the loop shape evolution is the increase in the magnetization. Thus the conclusions of this paper should be equally applicable to $ErFeO_3$ above the compensation point, where the magnetization increase is observed as well. As the temperature is raised starting from T_c, the magnetization grows all the way up to the spin-rotation transition temperature interval 88–97 K.[7,8] The few-domain theory thus would predict that a triangular temperature hysteresis should be also observed above T_c, while the argument based on the proximity to T_{N2} clearly would be inapplicable in this temperature range. Figure 5 shows the experimental results obtained on the $ErFeO_3$ sample No. 1. One can see well that at $H=20$ Oe and $H=30$ Oe there are two triangular hysteresis patterns, above and below T_c. Both of them occur when the magnetization reaches roughly the same value. The inset to Fig. 5 shows that the triangular-tail loops $M(H)$ occur in the temperature hysteresis region in the $T>T_c$ region. Similar to the case of $T<T_c$, it is the shape of these loops which determines the position of the magnetic moment jump during the temperature sweep. Similar measurements above T_c performed on the sample No. 3 are presented in Fig. 6. Here the hysteresis pattern at $T<T_c$ is overlapping with the butterfly pattern near T_c obscuring the picture, but the $T>T_c$ hysteresis is clearly

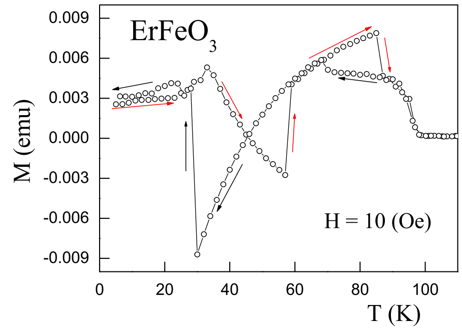

FIG. 6. (Color online) Magnetic moment of $ErFeO_3$ (sample No. 3) in a weak external field above and below T_c.

visible. Overall, these experiments prove that proximity to T_{N2} cannot be the cause of the triangular temperature hysteresis.

An additional way to test the few-domain theory is to notice that it provides a boundary for the external field H, above which the triangular temperature hysteresis should not be observed.[3] This critical field is proportional to the maximum value of M reached in the temperature sweep. Since the maximum value of magnetization for $T>T_c$ is smaller than for $T<T_c$ (see Fig. 1), the few-domain theory implies that above the compensation point the temperature hysteresis should be observed only at smaller values of external H. This is exactly the behavior observed in experiment. Below T_c the triangular hysteresis can be seen up to $H=200$ Oe, while above T_c it disappears already at $H>30$ Oe.

Finally, it is known that $TmFeO_3$ exhibits magnetization growth upon the temperature increase from 14 K up to the spin-rotation transition (82–92 K).[9] Figure 7 shows the $M(T)$ measured on the $TmFeO_3$ sample No. 4 in a small constant external field. The expected temperature hysteresis is indeed observed between 60 and 80 K where the magnetization reaches a high enough value. It is also worth noting that a comparison of Figs. 6 and 7 reaffirms the earlier conclusion[9] about the absence of a compensation point near $T=14$ K in $TmFeO_3$.

III. WEAK FERROMAGNETS BEHAVE LIKE NANOMAGNETS

Our magnetic measurements on the $ErFeO_3$ and $TmFeO_3$ single crystals support the picture of a two-domain ground state of a millimeter-size samples of weak ferromagnets.

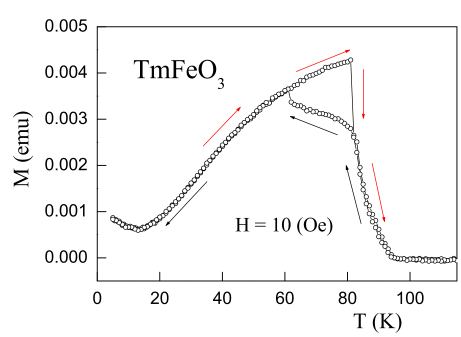

FIG. 7. (Color online) Temperature dependence of magnetic moment in $TmFeO_3$ (sample No. 4) in a weak external field $H=10$ Oe.

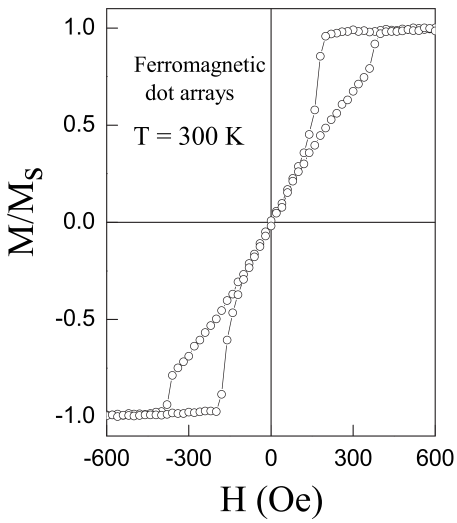

FIG. 8. Hysteresis loop of the square array of permalloy circular dots (dot diameter 1 μm, dot thickness 50 nm, center to center distance—1.2 μm, total patterned area 3×3 mm^2).

More generally, they underscore that the reason of the frequent dramatic differences in the properties of nanosize and massive samples is the fact that the dimensions of the nanosamples get close to one of the characteristic lengthscales of the material. The case of the weak ferromagnets shows that a few-domain magnetic state, normally associated with nanomagnets, can be actually achieved in much larger samples. In this case the relevant lengthscale is the magnetic length $\lambda_m = \sigma / 2\pi M_s^2$, where σ is the domain wall energy. This lengthscale diverges at $M_s \to 0$.

How weak should the ferromagnet be to have a few-domain structure in a millimeter range sample? The relationship between the characteristic length l_m and the sample size L that produces a few-domain state may be quite complicated, depends on the shape, and does not necessarily require $l_m \gtrsim L$. In Ref. 3 the case of a magnetic cube is discussed. Here $L < 25 l_m$ gives a single-domain sample, $25 l_m < L < 75 l_m$ corresponds to the presence of one domain wall, and further increase in L produces a progressively larger number of walls. Using these inequalities and setting σ to a representative value of 1 erg/cm^3, one finds that a $1 \times 1 \times 1$ mm^3 cube will have a one- or two-domain structure for $M_s \lesssim 10$ emu/cm^3. This estimate is unfortunately quite approximate due to the lack of good data on the domain wall energy. For $ErFeO_3$ it corresponds to a wide temperature interval of at least 15 K away from the compensation point.

It is worth comparing the field hysteresis loop shapes $M(H)$ in Figs. 2–4 with those found in submicron magnetic dots of strong ferromagnets.[10,11] Figure 8 shows an example of the hysteresis curve of a lattice of permalloy (Py) nanomagnets. The behavior of the Py disks and the orthoferrite rectangles is qualitatively similar in the most general sense of having a few discontinuous jumps that correspond to the magnetic defects entering or exiting the sample. The small number of defects is guaranteed by the large magnetic length in the material, as compared to the sample size. The details of the magnetic structure and the nature of magnetic defects in those two systems are of course different.[12] But since the Py disks and our orthoferrite samples both have just one defect, a hysteresis loop with two triangles connected by a central branch develops in both cases. This similarity stems directly from the low magnetization of orthoferrites and will be reproduced in the samples of any weak ferromagnets.

In contrast, the temperature transition manifesting itself in the triangular hysteresis of $M(T)$ (Figs. 1 and 7) requires an additional ingredient. Namely, the absolute value of magnetization M_s has to be significantly temperature dependent so that the sample would change its equilibrium magnetic structure with T. Weak ferrimagnets close to the compensation point are the natural candidates for systems with strong temperature dependence $M_s(T)$, but any other material with temperature dependent M_s will exhibit a similar behavior.

It should be mentioned that as any transition driven by the long range interactions (magnetic dipole interactions in this case), the triangular temperature hysteresis transition depends on the dimensions and shape of the sample. In this sense it can be viewed as a magnetic size effect.

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[12] In our particular example the permalloy and the orthoferrite differ qualitatively in the value of the factor $Q = 2\pi M_s^2 / K$, where K is the crystalline anisotropy constant. The opposite limits are realized for permalloy ($Q \gg 1$) and orthoferrite ($Q \ll 1$). This circumstance, together with the sample shape, leads to a sharp difference in the nature of magnetic defects: they are vortices in permalloy disks and domain walls in orthoferrite rectangles.