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Spin-Reorientation in $YbFeO_3$

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Precise measurements of $YbFeO_3$ magnetization in the spin-reoirentation temperature interval are performed. It is shown that ytterbium orthoferrite is well described by a modified mean field theory developed for $ErFeO_3$. This supports earlier conjectures about the importance of the rare earth ion's paramagnetism and it's anisotropy in all orthoferrites exhibiting $\Gamma_4 \to \Gamma_{24} \to \Gamma_2$ orientation phase transitions.

I. INTRODUCTION

Rhombic rare-earth orthoferrites $RFeO_3$ with R being a rare-earth ion or an yttrium ion are magnetic insulators that provide a classic example of the second order orientation phase transitions. Orthoferrites have of two magnetic subsystems: the one of the rare-earth ions, and the one of the iron ions. Magnetic properties of the subsystems and interaction between them depend on external parameters, e.g. the temperature, and a series of phase transition is observed upon the parameter change.

In the temperature interval where phase transitions discussed in this work take place, the iron subsystem is ordered into a slightly canted antiferromagnetic structure exhibiting a weak ferromagnetic moment **F**. The rare-earth system is paramagnetic. For all orthoferrites the

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antiferromagnetic structure below the Neel temperature T_N ($T_N = 620 \div 740$K) corresponds to the Γ_4 (G_x, F_z) irreducible representation with magnetic vector **F** pointing along the **c** axis of the crystal and antiferromagnetic vector **G** pointing along the **a** axis. The coordinates are chosen so that $\mathbf{c} = \hat{z}$ and $\mathbf{a} = \hat{x}$. In orthoferrites with non-magnetic rare-earth ions (R = La, Lu, or Y) the Γ_4 (G_x,F_z) configuration persists to the lowest temperatures. For many other orthoferrites a reorientation transition with the sequence Γ_4 (G_x,F_z) $\rightarrow$ Γ_{24} (G_{xz},F_{xz}) $\rightarrow$ Γ_2 (G_z,F_x) is observed. Upon cooling vector **F** starts to rotate away from the **c** axis at temperature T_1. Its continuous rotation towards the **a** axis happens in the $(\mathbf{a}, \mathbf{c})$ plane between temperatures T_1 and $T_2 < T_1$. Below T_2, the system stays in the Γ_2(G_z,F_x) phase with $F||\mathbf{a}$.

Although the spin reorientation region $[T_2, T_1]$ has been studied for many orthoferrites by different experimental techniques, not enough is known about the specifics of the rotation. Relevant experimental results are often incomplete, lack accuracy, tend to contradict each other, and do not correspond to either conventional Landau theory [1–3] or its modifications suggested by the authors. Recently [4, 5] the temperature dependence of both **a** and **c** axis projections of the magnetic moment was measured with high accuracy for the single crystal samples of $ErFeO_3$. These measurements gave the temperature dependence of the magnetization absolute value $M(T)$ and its rotation angle $\theta(T)$ with the respect to **c** axis in the $[T_2, T_1]$ temperature interval at zero external magnetic field. The results were in very good agreement with the proposed mean field model, [4] that emphasized the anisotropy of the rare-earth ions paramagnetic susceptibility. It was conjectured that this model would be suitable for other magnetic materials with similar phase transitions. The present study is aimed at the detailed measurements of the $M(T)$ and $\theta(T)$ behavior in single crystals of $YbFeO_3$, that exhibit the same $\Gamma_4 \rightarrow \Gamma_{24} \rightarrow \Gamma_2$ transition, with the purpose of checking this conjecture on another material. It is shown that the modified field theory of Refs. 4, 5 works well for $YbFeO_3$, even though in this orthoferrite the reorientation happens at an order of magnitude lower temperatures ($T \approx 8$K), than in $ErFeO_3$ ($T \approx 90$K), while the Neel temperature remains roughly the same $T_N \approx 630$K.

II. EXPERIMENTAL RESULTS

Measurements were performed on two single crystals of $YbFeO_3$. Sample A was a cube weighting 0.0485 g prepared by spontaneous crystallization in the melt-solution. Sample B was an ellipsoid weighting 0.0715 g grown by the no-crucible zone melting technique with radiation heating. The results for both samples are very similar. The temperature was varied in the $2 \div 10$K interval, and both M_a and M_c projections of the magnetic moment were measured by a Quantum Design MPMS-5S SQUID magnetometer.

The $\mathbf{M}(T)$ dependence at zero external magnetic field was found through the analysis of magnetization curves analogous to those shown in Fig. 1. The $(H-T)$ phase diagrams in the vicinity of the $\Gamma_4 \rightarrow \Gamma_{24} \rightarrow \Gamma_2$ transition for $\mathbf{H}||\mathbf{c}$ and $\mathbf{H}||\mathbf{a}$ field directions are well known. They show, that when magnetic field is swept through $H = 0$ inside the $[T_2, T_1]$ reorientation interval, a first order transition in at least one magnetization component happens for both directions of the field. First order transitions also happen above T_1 for $\mathbf{H}||\mathbf{c}$ and below T_2 for $\mathbf{H}||\mathbf{a}$ orientations, while no transitions are predicted below T_2 for $H||\mathbf{c}$ and above T_1 for $H||\mathbf{a}$. In a real experiment exact orientation of the field direction is obviously impossible. A three-dimensional diagram valid for the arbitrary field direction [4, 6] shows that for a tilted field a first order transition happens at any temperature and a jump of at least one magnetic moment projection should be observed with a rectangular hysteresis loop in the case of single-domain switching.

Well-developed rectangular loops were indeed observed in experiments on $ErFeO_3$ outside of the reorientation interval. [4] Inside the $[T_2, T_1]$ interval ($T_1 \approx 88$K and $T_2 \approx 97$K for erbium orthoferrite) they transformed into the S-shaped magnetization curves. Such modification was attributed to the multi-domain state formation, possibly connected to the abrupt change in domain wall mobility. [7]

In contrast with the case of $ErFeO_3$, magnetization curves in $YbFeO_3$ are S-shaped at all temperatures studied here for both $\mathbf{H}||\mathbf{a}$ and $\mathbf{H}||\mathbf{c}$ field orientations. A small hysteresis was observed only in the reorientation region. The width of the magnetization curves for the magnetic field directed along the **a** axis is larger then for the field along the **c** axis. In general, the total width of the loops is considerably larger than in the case of $ErFeO_3$. [4] In accord with the phase diagrams discussed above, magnetization curves become straight lines passing through the origin above T_1 for the $\mathbf{H}||\mathbf{a}$ orientation and below T_2 for the $\mathbf{H}||\mathbf{c}$

orientation. Their slope corresponds to the paramagnetic contribution of the ytterbium ions. Importantly, magnetization curves obtained for different samples are very similar.

The shape of the magnetization curves and the presence or absence of hysteretic behavior depends on the quality of the samples, energy of the domain walls, etc. The observed difference between erbium and ytterbium orthoferrites may result from the order of magnitude difference in the temperature of the reorientation transition. This question requires a separate study. Here we simply extract the **a** and **c** projections of the bulk magnetization at zero external field by extrapolating their observed linear dependence at higher fields, $H \gtrsim 650$ Oe, by $M_{a,c}(H,T) = M_{a,c}(T) + \chi_{a,c}(T)H$ and extracting the vertical intercept $M_{a,c}(T)$. The values of $M_a(T)$ and $M_c(T)$ obtained through this procedure are shown in Fig. 2. The absolute value of magnetization M and rotation angle θ are calculated from the expressions

$$M = \sqrt{M_a^2 + M_c^2}\,, \quad \theta = \arctan\left(\frac{M_a}{M_c}\right)$$

and are shown in Fig. 3 and Fig. 4. Experimental results presented in Figs. 2-4 show that as the temperature is lowered from T_N to T_1, the magnetization of the crystal gradually grows. This shows the build up of iron moment near T_N and developemnt of ytterbium moment along iron moment [8] at lower temperatures. In the narrow reorientation region $[T_2, T_1]$ the magnetization rapidly grows almost two-fold. Below T_2 the magnetization continues to grow. This suggests that ytterbium moment remains parallel to the iron moment, and does not switch to the antiparallel direction as stated in Ref. 8.

III. THEORETICAL ANALYSIS

Our experimental results can be explained by the modified mean field theory suggested in Refs. 4, 5. Analogous to the conventional Landau theory, [1–3] the modified theory assumes that the magnetization of iron subsystem is saturated at $T \lesssim T_{1,2} \ll T_N$. The free energy of the iron subsystem is taken in the form

$$F(\theta,T) = F_0(T) + \frac{K_u(T)}{2}\cos(2\theta) + K_b\cos(4\theta) \tag{1}$$

With minimal assumptions about the temperature dependence of phenomenological constants inside the reorientation region, namely constant K_b, and $K_u(T)$ linearly varying with

temperature and going through zero inside the reorientation interval, the minimization of the conventional energy functional (1) gives [1–3]

$$\tan\theta \;=\; \sqrt{\frac{1+\xi}{1-\xi}}\;, \quad \xi(T) = \frac{(T_1+T_2)/2 - T}{(T_1-T_2)/2} \tag{2}$$

Figs. 3, 4 show that experimental results neither support the constancy of $M(T)$, nor give a $\theta(T)$ dependence consistent with Eq. (2)

According to the modified mean field model, paramagnetic susceptibility of ytterbium subsystem should also be taken into account to adequately describe the magnetic behavior of the orthoferrite. It is assumed, that in the molecular filed of iron the rare-earth ion acquires magnetic moment $\mathbf{m} = \hat{\chi}^{\mathrm{Yb}}\mathbf{F}$, while the absolute value of the iron moment $\mathbf{F}$ is indeed constant. [2, 8–10] Experimentally measured magnetization is a sum of iron and rare-earth contributions $\mathbf{M} = \mathbf{F} + \mathbf{m}$. Magnetic susceptibility $\hat{\chi}^{\mathrm{Yb}}$ of the rare-earth ions is assumed to be anisotropic. This assumption naturally explains the large change of M inside a narrow temperature interval, since rotation of $\mathbf{F}$ leads to the change of $\mathbf{m}$ and thus changes M as well.[4] The anisotropy of rare-earth susceptibility was discussed in the literature. [2, 8–10] The key point of Ref. 4 was the proper account of it in the calculation of the temperature dependence of the rotation angle and absolute value of magnetization, with the result:

$$\tan\theta \;=\; r\sqrt{\frac{1+\xi}{1-\xi}}\;, \quad r = \frac{M_a(T_2)}{M_c(T_1)} \tag{3}$$

$$M \;=\; M_c(T_1)\sqrt{\frac{r^2(1+\xi)+(1-\xi)}{2}} \tag{4}$$

Since $M_a(T_2)$ and $M_c(T_1)$ are measurable magnetizations of the sample at temperatures T_2 and T_1 respectively, the value of r is known and expressions (3) and (4) have no fitting parameters. Note that they are only valid for temperatures inside the reorientation region $[T_2, T_1]$.

According to our measurements, the cubic sample A, for which most of the measurements were done, had $T_1 = 8.0$ K and $T_2 = 6.6$ K. Using the values of $M_a(T_2)$ and $M_c(T_1)$ at these temperatures we find $r = 1.78$. Theoretical curves given by Eqs. (3) and (4) are shown in Figs. 3 and 4 by solid lines. A convincing correspondence between the theory and experiment is evident and serves as a strong argument if favor of the spin reorientation model suggested

in Ref. 4 and its applicability to Γ_4 $(G_x,F_z) \rightarrow \Gamma_{24}$ $(G_{xz},F_{xz}) \rightarrow \Gamma_2$ (G_z,F_x) orientation transitions in different materials.

IV. ACKNOWLEDGEMENTS

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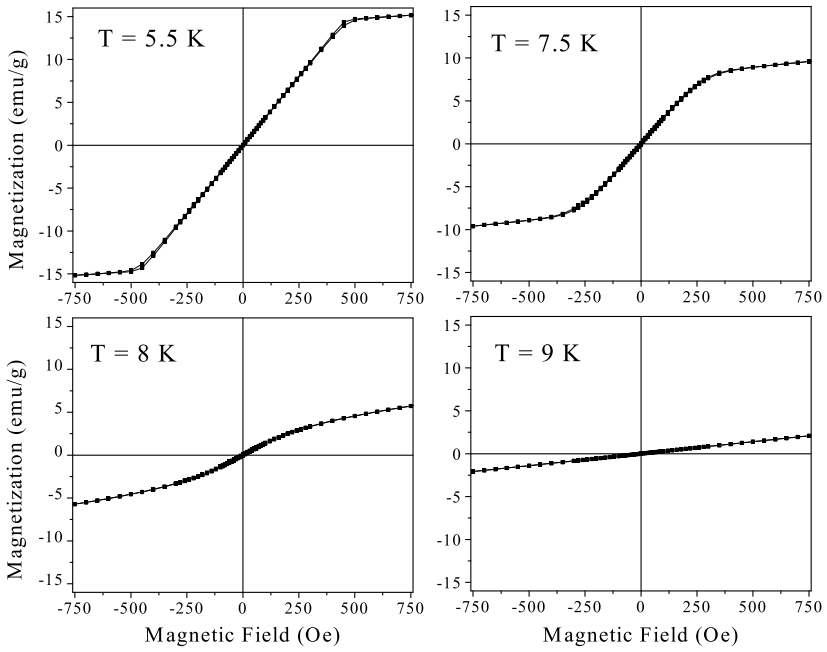


FIG. 1: Examples of magnetization curves $M_a(H)$ obtained on the $YbFeO_3$, sample B, at different temperatures.

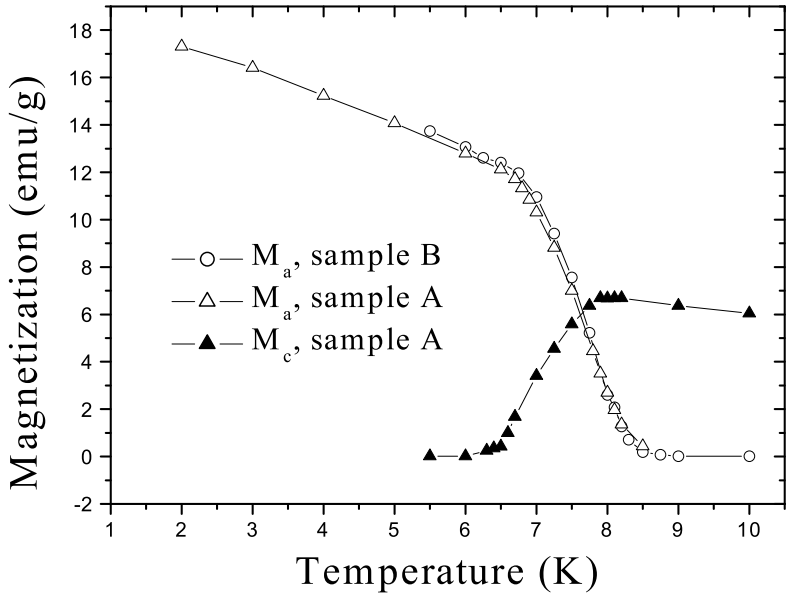


FIG. 2: Magnetization projections $M_{a,c}(T)$ obtained from the magnetization curves: empty circles - $M_a(T)$ for sample B, empty triangles - $M_a(T)$ for sample A, filled triangles - $M_c(T)$ for sample A.

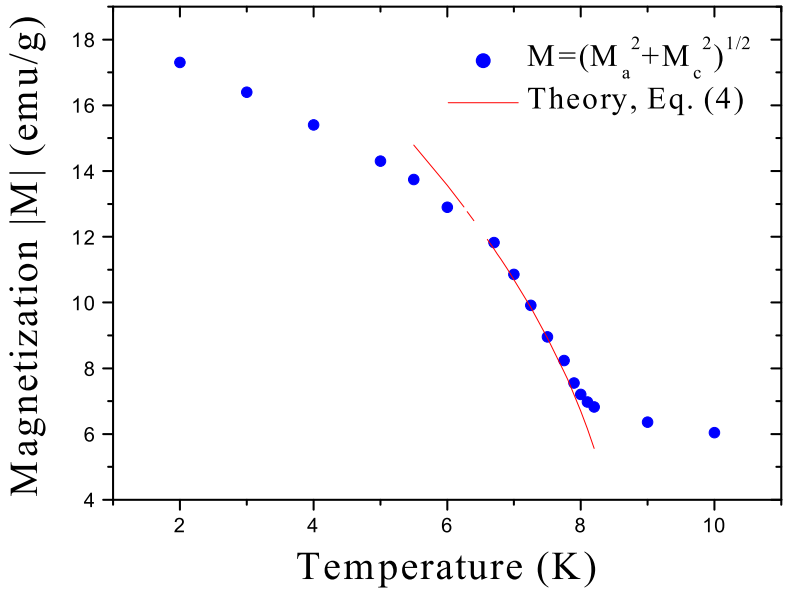


FIG. 3: Absolute value of the magnetization $M(T)$ calculated from experimental data. Solid curve - theory Eq. (4)

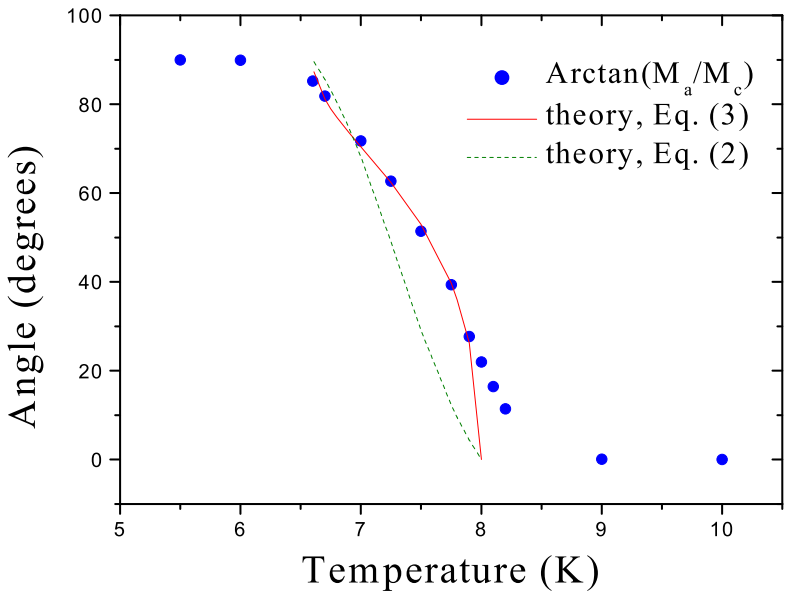
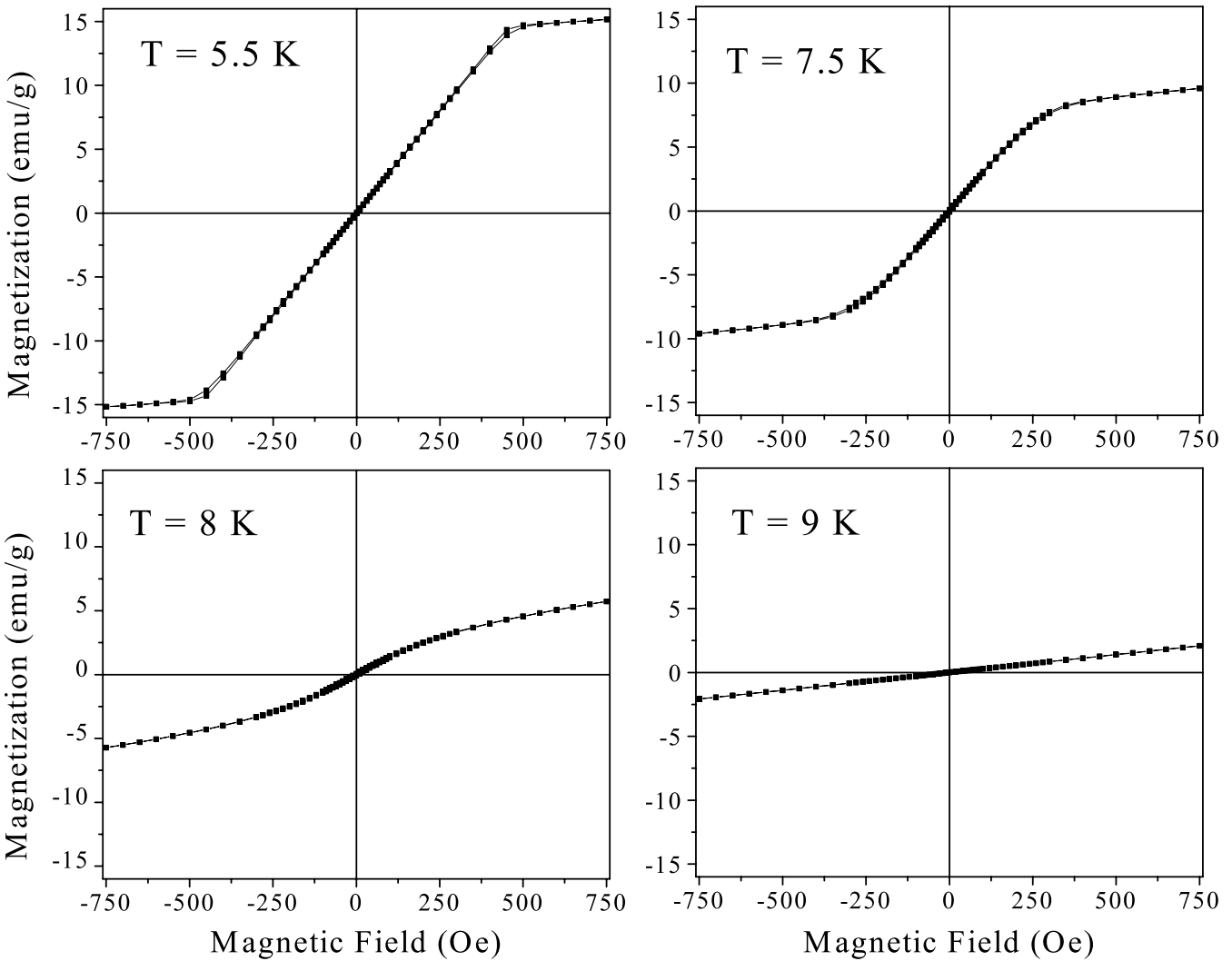
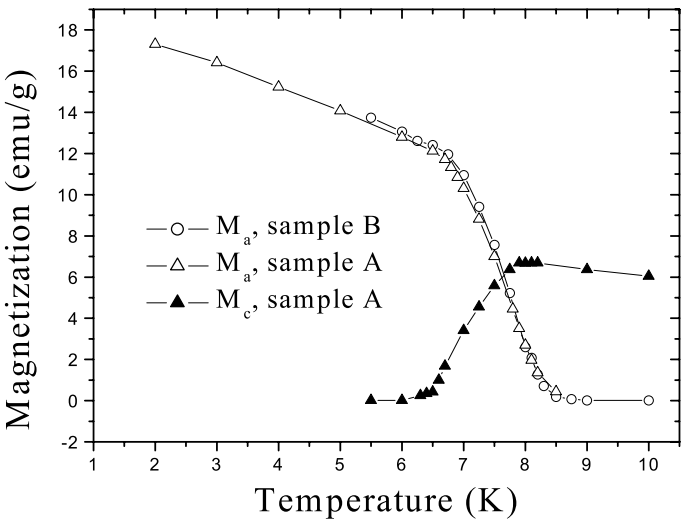
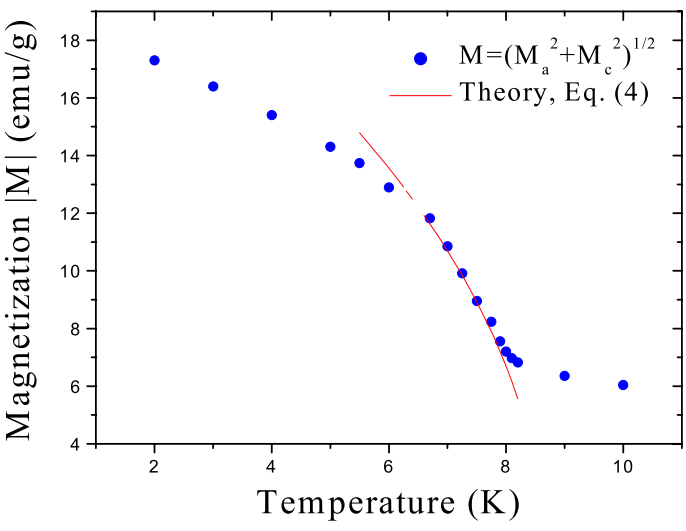
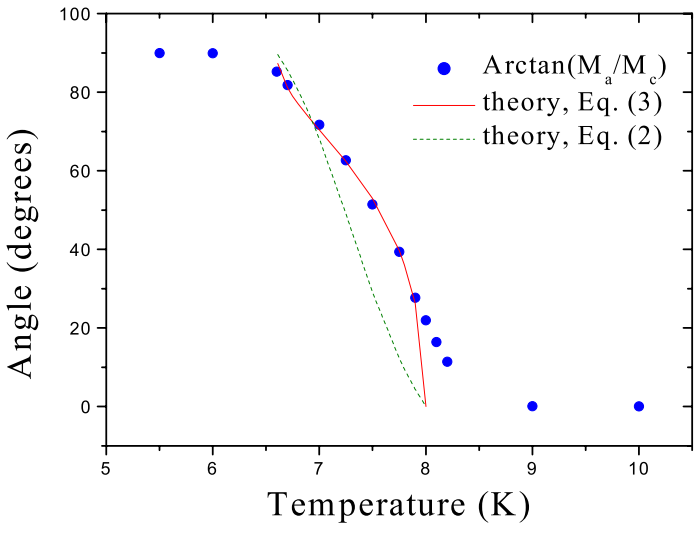


FIG. 4: Magnetization rotation angle $\theta(T)$ calculated from experimental data in the reorientation region $[T_2, T_1]$ at zero external magnetic field. Solid curve - theory Eq. (3), dash curve - conventional theory Eq. (2)


T = 5.5 K
T = 7.5 K
T = 8 K
T = 9 K
Magnetization (emu/g)
Magnetization (emu/g)
Magnetic Field (Oe)
Magnetic Field (Oe)
15
10
5
0
-5
-10
-15
-750
-500
-250
0
250
500
750


Magnetization (emu/g)
Temperature (K)
M_a, sample B
M_a, sample A
M_c, sample A
18
16
14
12
10
8
6
4
2
0
-2
1
2
3
4
5
6
7
8
9
10


Magnetization |M| (emu/g)
Temperature (K)
M=(M_a^2+M_c^2)^1/2
Theory, Eq. (4)
18
16
14
12
10
8
6
4
2
4
6
8
10


Angle (degrees)
Temperature (K)
Arctan(M_a/M_c)
theory, Eq. (3)
theory, Eq. (2)
100
80
60
40
20
0
5
6
7
8
9
10