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M. Marysko, Z. Frait, S. Krupicka. FMR and Static Magnetic Properties of Gallium Substituted Magnetoplumbite. Journal de Physique IV Proceedings, 1997, 07 (C1), pp.C1-347-C1-348. 10.1051/jp4:19971140 . jpa-00254784

HAL Id: jpa-00254784

<https://hal.science/jpa-00254784>

Submitted on 1 Jan 1997

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FMR and Static Magnetic Properties of Gallium Substituted Magnetoplumbite

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Abstract. Magnetization and uniaxial anisotropy were studied on single crystals of $\text{PbFe}_{12-x}\text{Ga}_x\text{O}_{19}$, where $x = 0 - 8$ from 2 to 360 K. The results suggest that for $x > 3$ the spin arrangement becomes non-collinear. The magnetic moment and anisotropy for $x < 3$ are roughly consistent with the random distribution of Ga^{3+} ions in all sites except for 2b. For $x = 5.94$ and 8.02 the relatively large anisotropy of the Curie temperature was found. The FMR measurements at $T = 300$ K were interpreted using the g-tensor formulation.

1. INTRODUCTION

We are concerned with the single crystals of the magnetoplumbite $\text{PbFe}_{12-x}\text{Ga}_x\text{O}_{19}$ substituted by gallium. Although the preparation and some properties of these materials have been reported earlier [1-3], detailed work on this subject was absent. In addition to it the first FMR experiments made for $x \leq 3.1$ did not allow to make the definite conclusions on this system. The purpose of this article is to report the static as well as the FMR measurements performed on additional samples especially, with larger Ga content.

2. EXPERIMENT AND RESULTS

The single crystals of $\text{PbFe}_{12-x}\text{Ga}_x\text{O}_{19}$ (for x see Tab. 1) were prepared by the flux method [3]. The samples used in experiments were platelets of about square form with the transversal dimensions 1.5 - 2 mm and thickness 50 - 150 μm . They were obtained by splitting and polishing so that the surface of the platelet was perpendicular to the hexagonal c axis.

The static measurements were performed by the magnetometer Quantum Design MPMS-5S. We measured the magnetization curves in easy and hard directions i.e. with the static magnetic field along ($\parallel$) and perpendicular ($\perp$) to the c axis respectively. The temperature range was from $T = 2$ to 360 (400) K. In the field region 30 - 50 kOe the susceptibility of the paraprocess χ_p was evaluated. At $T = 2$ K we find $(\chi_p)_{\parallel} \approx (\chi_p)_{\perp} < 10^{-5}$ for $x=0$; 1.21 and $2.3 \cdot 10^{-5}$; $3.34 \cdot 10^{-5}$; $1.07 \cdot 10^{-4}$; $1.72 \cdot 10^{-4}$; $3 \cdot 10^{-4}$ for $x=3.1$; 4.49; 5.94; 8.02 respectively. For $x \leq 3.1$ χ_p increases markedly with increasing temperature whereas for $x \geq 4.49$ χ_p becomes nearly temperature independent. The saturation magnetization M_s was determined by extrapolating the magnetization curves to $H=0$ and for $T > 0.85 T_c$ using the Arrot plot M^2 vs M/H . For $H > 15$ kOe this plot corresponding to the ($\parallel$) and ($\perp$) direction was linear and we could evaluate $(M_s)_{\parallel}$ (Fig. 1 a), $(M_s)_{\perp}$ and the Curie temperatures $(T_c)_{\parallel}$, $(T_c)_{\perp}$ in the standard way. For $x=4.49$ we find $(T_c)_{\parallel} \approx (T_c)_{\perp} \approx 416.5 \pm 0.5$ K by extrapolating from the region $T \leq 400$ K. In contrast to it, for $x = 5.94$ and 8.02 we get the anisotropic Curie temperature, namely $(T_c)_{\parallel} \approx 322 \pm 2$ K, $(T_c)_{\perp} \approx 312 \pm 2.5$ K for $x = 5.94$ and $(T_c)_{\parallel} \approx 127 \pm 5$ K, $(T_c)_{\perp} \approx 104 \pm 2$ K for $x = 8.02$. A similar behaviour is observed for the paramagnetic Curie temperatures determined from the $1/\chi$ vs T plots. The uniaxial anisotropy fields were evaluated from the slopes s of the ($\perp$) magnetization curves at $H=0$ using the relation $2 K_1/M_s = M_s/s$. (Fig. 1 b).

The ($\parallel$) and ($\perp$) FMR measurements were performed at frequencies 46.6; 59.5; 69.5; 79.5 and 50.6; 59 GHz respectively. From the measured resonance fields we evaluated the components of the g tensor $g_{\parallel}$, $g_{\perp}$ and the effective anisotropy fields H_1^{eff} , H_2^{eff} , where $H_1^{\text{eff}} = 2K_1/M_s - 4\pi M_s$ [4]. For $x \leq 4.49$ the peak-to-peak FMR linewidth ΔH_1 varies from about 100 Oe (46 GHz) to 150-250 Oe (69 GHz), the corresponding ΔH_1 values being roughly two times larger. For $x = 5.94$ we obtain $\Delta H_1 \approx 800$ Oe which is relatively independent on frequency ($\Delta H_1 \approx 600$ Oe).

3. DISCUSSION

The behaviour of χ_p suggests that for $x > 3$ the arrangement of the spins becomes non-collinear and therefore we limit ourself, in the quantitative considerations, to the region $x < 3$. Here the dependence of the measured magnetic moment n and constant K_1 on x (Fig. 1 b) at $T = 2$ K will be compared with the theoretical functions $n(x)$, $K_1(x)$ (the single-ion contributions ΔK_1 , taken from [5] and $n(0)$, $K_1(0)$ fitted to the experiment).

Figure 1: a) Temperature dependence of the saturation magnetization $(M_s)_I$ for $x=0(O) \div 8.02(+)$.

b) Measurement at $T=2$ K - magnetic moment per formula unit (O), K_I (■) as functions of x ; straight lines correspond to different distributions of Ga^{3+} ions.

Tab. 1. Data at $T = 300$ K.

x	$g_{\parallel}$	$g_{\perp}$	$H_1^{eff}(Oe)$	$H_{\perp}^{eff}(Oe)$	$4\pi M_s(G)$	$2K_I/M(Oe)$	$(H_{\perp}^{eff})_s(Oe)$
0	1.982	1.959	9794	9135	4499	13930	9431
1.21	1.982	1.954	11513	11038	3908	14739	10831
2.19	1.915	1.955	14664	13220	3104	16209	13105
3.1	1.906	1.986	15921	15340	2450	17727	15277
4.49	2.000	1.976	13126	13060	1621	16797	15176
5.94	1.914	1.912	13287	12526	447	11374	10927

For the distribution of the Ga^{3+} ions among 12 k and 2 a sites [6], among all the sites and among all the sites, except for the 2 b sites [7] we obtain the dotted, dashed-dotted and dashed straight lines respectively. From these three distributions relatively the best agreement occurs for the latter case.

We regard as an interesting fact the relatively large anisotropy of the Curie temperature (ΔT_c up to 23 K) observed for $x=5.94$ and 8.02. A similar but smaller effect of this kind was reported for $PbFe_{12}O_{19}$ [8] ($\Delta T_c \approx 1.2$ K) and $BaFe_{12}O_{19}$ [9] ($\Delta T_c \approx 3.8$ K). For a ferromagnet the anisotropy of T_c should appear for the ratio λ of the anisotropy to exchange energy comparable to unity [10], which is not our case ($\lambda \approx 10^{-2}$). Abrupt increase of ΔT_c when going from $x=4.49$ to 5.94 suggests, however, that the observed effect may be, in a way, connected with increasing disordering of the magnetic system for the large dilution by nonmagnetic Ga^{3+} ions. This conjecture is also supported by the fact that an unexpected increase of ΔH was found just for $x=5.94$.

For $x \leq 3.1$ the fields H_1^{eff} , $H_{\perp}^{eff}$ agree relatively well with the values of $(H_{\perp}^{eff})_s$ which are calculated from $2K_I/M_s$ and $4\pi M_s$ (Tab. 1). In accord with [4] this agreement is better just for H_1^{eff} . The disagreement for $x=4.49$; 5.94 is probably connected with a non-collinear arrangement. Analogously as for $BaFe_{12-x}Ga_xO_{19}$ [11] we find a frequency independent part of ΔH due to inhomogeneities. For $x=5.94$, where M_s is small, the large value of this contribution is likely to be caused by fluctuations in K_I and $g_{\parallel}$, $g_{\perp}$.

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