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Abstract. – The low temperature behaviour of the magnetization and magnetic anisotropy was studied on epitaxial films Ca:YIG. The results were discussed using a new model in which the main role is played by the strong exchange interaction between the paramagnetic O^- and neighbouring Fe^{3+} ions.

1. Introduction

Recently a considerable attention has been given to the study of CaGe:YIG, Ca:YIG, CaGa:YIG epitaxial films in connection with the observed low temperature magnetization anomalies [1-4]. These effects were explained on the basis of the assumption that towards lower temperatures the hole is gradually localized and forms the tetrahedral Fe^{4+} (d) ion, which is ferromagnetically coupled to the octahedral Fe^{3+} (a) ions. (According to the Goodenough-Kanamori rules we would rather expect the antiferromagnetic interaction.) The purpose of the present work is to bring further information on the simple system Ca:YIG and to discuss possible explanations.

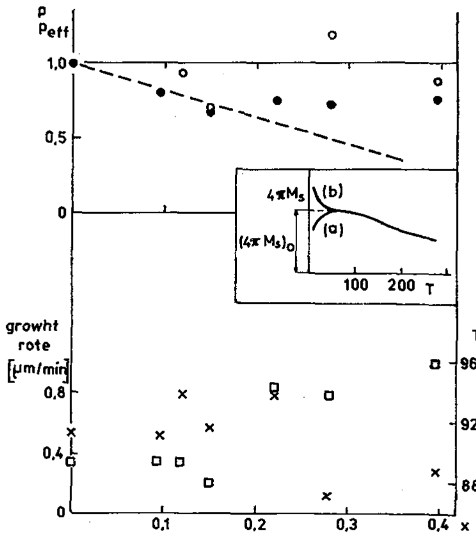

Fig. 1. – Growth rate (×), growth temperature (□), ratio p (○), ratio p_{eff} (•) for seven films having the different Ca content; the dashed line corresponds to the function $p = 1 - 9x/5$. In the inset, the sketch of the typical measured dependence $4\pi M_s(T)$.

2. Experimental procedure and results

A series of garnet expitaxial films of composition $Y_{3-x-y}Ca_xPb_yFe_{5-z}Pt_zO_{12}$ with x ranging from 0 to 0.39, $y < 0.028$, $z < 0.03$ and the thickness between 1.8 and 8 μm was investigated. The samples were grown by the LPE method (growth rates and temperatures see Fig. 1) on the (100) and (111) oriented substrates and the final compositions were determined by the EPMA. The effective magnetization $4\pi M_{eff} = 4\pi M_s - 2K_u / M_s$ and the cubic anisotropy field $2K_1 / M_s$ were evaluated from the FMR measurements at 25 GHz between $T \approx 7$ and 300 K. For some samples the saturation magnetization $4\pi M_s$ was measured using a vibrating sample magnetometer. Two types of the measured dependence $4\pi M_s(T)$ are shown in figure 1 (it also holds for $4\pi M_{eff}$). For the sample with $x = 0.28$ and the smallest growth rate we find the type (b) exhibiting an increase of $4\pi M_s$ at $T < 40$ K; for the other films and in all cases for $4\pi M_{eff}$ the type (a) with a decrease of $4\pi M_s$ at $T < 30 - 50$ K is seen. To characterize this low temperature behaviour, we introduce the quantities $p = 4\pi M_s(0) / (4\pi M_s)_0$ and $p_{eff} = 4\pi M_{eff}(0) / (4\pi M_{eff})_0$ which are determined by extrapolating to $T = 0$ K. In the upper part of figure 1 the ratios p and p_{eff} are plotted *vs.* Ca content. For small x the ratio decreases with increasing x and then we see a minimum or a region of the saturation. For all films in the measured temperature region the contribution $\Delta K_1 = K_1 - K_1$ (YIG) is small; namely $|\Delta K_1| < 1.5 \times 10^4$ erg/cm^3, which corresponds to $|\Delta K_1| < 0.1$ cm^{-1} per ion of the dopant. The uniaxial anisotropy constant K_u at $T < 30$ K increases and its value extrapolated to $T = 0$ K is about 10^4 erg/cm^3 $K_u(0) \approx (p - p_{eff}) . (4\pi M_s/2)$).

3. Discussion

Our experimental results agree with those in [3, 4] and supplement them namely as far as the dependence on Ca content and the observation of a low temperature increase of $4\pi M_s$ is concerned. As in [3] we come to the conclusion that K_1 is much smaller than that expected for the Fe^{4+} (d) ions. Especially the latter fact but also a peculiar dependence of p on Ca content do not support the Fe^{4+} (d) model, which yields $p = 1 - 9v/5$, where v is the concentration of the Fe^{4+} (d) ions. In [5] the charge compensation was ascribed to holes on O^{2-} ions. We may assume that towards lower temperatures these holes become gradually localized forming the O^- ions. We now show that the magnetization anomalies can be alternatively explained by strong exchange interactions between the paramagnetic O^- and neighbouring Fe^{3+} ions. Similar mechanism has been recently suggested for the configuration $Cu^{2+}-O^- -Cu^{2+}$ occuring in the high temperature superconductors [6].

It is reasonable to assume that the magnitude of exchange integral J, which characterizes $Fe^{3+}-O^-$ interaction is much larger than the integral of Fe^{3+} (d) – Fe^{3+} (a) superexchange. As a consequence the Fe^{3+} (d) and Fe^{3+} (a) spins will be parallel to each other regardless of the sign of J. Since in YIG the Fe^{3+} (d) and Fe^{3+} (a) spins are antiparallel, it means that either the Fe^{3+} (d) or Fe^{3+} (a) spin will be reversed. For an isolated O^- ion (i.e. small Ca concentration) the reversion will take place at the Fe^{3+} (d) spin at which the molecular field is smaller. Thus the magnetic moment p.f.u. will decrease by 9 or 11 μ_B depending on the orientations of the O^- spin. The corresponding contribution Δ to $4\pi M_s$ is given by $\Delta = -(9w/5)\,4M_s$ and $\Delta = -(11w/5)4M_s$ for $J < 0$ and $J > 0$ respectively. Here w denotes the concentration of the O^- ions. For larger Ca content the situation is more complex since we must consider configurations in which e.g. two ions O^- will be close together. For electrostatic reasons the configuration $O^- -Fe^{3+}$ (a) $-O^-$ will be probably preferred. In this case, as can be shown, the Fe^{3+} (a) spin will reverse, which (for $J < 0$) corresponds to $\Delta = (9w/10)\,4\pi M_s$. The quantitative analysis is complicated by the fact that the Ca^{2+} ions and thus the O^- ions may also form clusters in which their concentration is larger that on the average. In summary, for small w the suggested mechanism yields $p = 1 - 9w/5(J < 0)$ and for larger w we may expect the decrease of the slope $|dp/\,dw|$ or the occurence of a minimum. This could correspond to the average course of the measured dependence $p = p\,(x)$ if we put $w \approx x$.

The more detailed inspection of figure 1 shows e.g. that for $x = 0.15$ the measured value of p corresponds to $p = 1 - 9w/5$, where $w \approx x$. The charge compensation could be thus attributed almost entirely to the O^- ions. Another situation occurs, however, for the film with $x = 0.12$; $y = 0.017$ where the measured value of p yields $w \approx 0.035$. Here, the remaining concentration $x - y \approx 0.07$ of Ca^{2+} ions must be compensated in the different way, probably by creation of Fe^{4+} (d) ions in the strong field configuration. The presence of a small concentration of the Fe^{4+} (d) ions ($S = 2$) cannot be however excluded. The number of the O^- or Fe^{4+} ions participating in the compensation mechanism does not appear to be only a function of the Ca content but depends also on the growth conditions. If it is assumed that the concentration of Pt remains small this result can be hardly understood. An explanation could be connected with an inhomogeneous distribution of Ca within the film for which, in the framework of our O^- model, the simple relation $p = 1 - 9w/5$ should not be used to estimate the value of w.

[1] Wigen, P. E. and Parrdavi-Horvath, M., Physics of magnetic materials (world scientific, Singapore) 1987, p. 91.

[2] Batra, S., Lehman-Szweykowska, A. and Wigen, P. E., *J. Appl. phys.* **61** (1987) 3274.

[3] Pardavi-Horvath, M., Wigen, P. E., Vertesy, G. and Degasperis, P., *IEEE Trans.* **MAG. 23** (1987) 3730.

[4] Pardavi-Horvath, M., Thavendrarajah, A. and Wigen, P. E. tto be published in *J. Appl. Phys.* **63** (1987) 3107.

[5] Roode, W. H. and Van de Pavert, C. A. P. W., *J. Appl. Phys.* **55** (1984) 3115.

[6] Aharony, A., Birgenau, R. J., Coniglio, A. and Kastner, M. A., Stanley, H. E., *Phys. Rev. Let.* **60** (1988) 1330.