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Magneto-optical Properties of Ce-Doped YIG Single Crystals

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Abstract. The Faraday rotation (FR) of cerium substituted yttrium iron garnets single crystals $Y_{3-x}Ce_xFe_5O_{12}$ (YIG:Ce) with $x=0.055$ has been investigated at 1152 and 632.8 nm in the 6 K - 600 K temperature range and magnetic field up to 20 kOe applied along the [111] and [100] directions. In contrast to magnetic properties, the FR of YIG:Ce is strongly different from FR of pure YIG at both wavelengths. The FR contribution of the Ce^{3+} ions, mainly attributed to intra-ionic electric dipole transitions between the $4f^1$ and $4f^0 5d^1$ configurations, is discussed within the frame of the one-ion model of ferrimagnetic garnets.

1 - INTRODUCTION

Recently, it has been shown that Ce-substituted yttrium-iron-garnet (YIG:Ce) are very promising magneto-optical (MO) materials both for MO applications and fundamental studies[1-6]. This is due to the specific efficiency of the Ce^{3+} ions which is much larger than that of Bi^{3+} ions. Much work deal with the wavelength dependence, at room temperature and fixed magnetic field, of the MO properties of these materials essentially as thin films. We present here the results of the Faraday rotation (FR) of YIG:Ce single crystals at two fixed wavelengths: 1152 nm (infra red) and 632.8 nm (visible) as a function of temperature and magnetic field and discuss briefly these results within the frame of the one-ion model[7].

2 - EXPERIMENTAL

Garnet single crystals of composition $Y_{3-x}Ce_xFe_5O_{12}$ were synthesized by flux growth. Polished oriented platelets were obtained from the "as grown" single crystals. The cerium concentration $x=0.055$, closed to its limit of substitution in YIG garnet synthesized by this method, was measured on the platelets by electron microprobe analysis and checked further by elemental chemical analysis. A small amount of lead was equally incorporated in the crystals ($x < 0.01$) but no platinum was detected. FR measurements were performed at $\lambda = 1152$ nm and 632.8 nm wavelengths in the 6 K - 600 K temperature range under a magnetic field up to 20 kOe applied parallel to the crystallographic axes [100] and [111]. The FR isotherms $\Phi(H, \lambda)$ present a linear part when $H \gg 6$ kOe and the spontaneous FR Φ_s is obtained by extrapolating to $H=0$ using the relation: $\Phi = \Phi_s + \chi_F H$, where χ_F is the FR susceptibility. Magnetization and Curie temperature T_C measurements were performed on the same crystals at temperatures between 1.5 and 600 K using a MANICS DSM10 susceptometer.

3 - RESULTS AND DISCUSSION

Cerium is incorporated as paramagnetic Ce^{3+} and diamagnetic Ce^{4+} ions[2],[5]. Assuming that the Pb^{2+} ions provide the electric charge compensation, the Ce^{3+} ions substitution rate is $x=0.045$ in the YIG:Ce crystals. The measured magnetic properties of the crystals are very close to those of pure YIG: the Curie temperature deduced from the temperature variation of M_s^2 in the vicinity of T_C is the same (560 K) and furthermore, at 1.5 K and 300 K the magnetization of YIG:Ce (Fig.1) differs from that of pure YIG by at most 4%. This low difference hinders the determination of the dodecahedral site magnetization of the Ce^{3+} ions from the magnetic properties as in other rare earth iron garnets[8].

On the contrary, notwithstanding the low Ce substitution rate, a strong enhancement of the FR by comparison to pure YIG is observed at both wavelengths. In the IR at 1152 nm, the FR is negative in the whole temperature range whereas the FR of pure YIG is positive (Fig. 1). At 632.8 nm wavelength the FR is positive like FR of pure YIG[9] but of very different magnitude particularly at low temperatures (Fig. 2). It is to be noted that our results are close to those obtained on LPE thin

Fig. 1 - Temperature dependence of Φ_s at 1152 nm
Inset: Magnetization isotherms at 1.5 K and 295 K

Fig. 2 - Temperature dependence of Φ_s at 632.8 nm
Inset: Temperature dependence of χ_F (1152 and 632.8 nm)

films by Kucera et al.[5] at the two temperatures 8K and 300 K with the exception of the Ce substitution rate which was probably underestimated[3]. The χ_F values are negative and quasi constant in the whole temperature range, except near T_C , for both wavelengths and nearly twice in magnitude at 1152 nm wavelength as compared to pure YIG (Fig. 2). Below 100 K an anisotropy of the Φ_s values is observed for both 1152 and 632.8 nm wavelengths with [100] being the "hard" axis. This FR anisotropy may be due, at least partially, to the magnetic anisotropy of these compounds as evidenced by ferromagnetic resonance method[10].

Both the Ce and Fe sublattices contribute to the FR. Within the frame of the one-ion model of Crossley et al.[7], the total contribution of the Fe^{3+} sublattices can be well represented by the FR of pure YIG[8]. The FR contribution of the Ce^{3+} ions in dodecahedral {c} sublattice, mainly attributed to intra-ionic electric dipole transitions between the $4f^1$ and $4f^0 5d^1$ configurations, is deduced by subtracting the FR of pure YIG from the experimental FR of YIG:Ce (Fig. 3). At 300 K, the specific cerium FR is $-1.1 \cdot 10^4$ and $1.5 \cdot 10^4$ °/cm/ Ce^{3+} at respectively 1152 and 632.8 nm. The Ce^{3+} contribution is given by [7], [8]: $\Phi_c(T, \lambda) = [C_m + C_e(T, \lambda)] |M_c(T)|$ where C_m and C_e are the MO coefficients due to magnetic(m) and electric(e) dipole transitions. The temperature variation of the reduced FR $\Phi_c(T, \lambda)/\Phi_c(0, \lambda)$ is plotted in Fig.3 for both wavelengths. When $T < 350$ K, the two curves are nearly the same within experimental accuracy indicating a similar temperature dependence of the reduced MO coefficient $[C_m + C_e(T, \lambda)]/[C_m + C_e(0, \lambda)]$ at these wavelengths. In the absence of experimental values, the reduced magnetization can be calculated using

Fig. 3 - Temperature dependence of the reduced FR of the Ce^{3+} ions.
Inset: Reduced MO coefficient and calculated $C_c(T, \lambda)/C_c(50, \lambda)$ [3]

$M_c(T)/M_c(0) = \tanh(\Delta/2kT)$ where $\Delta = 200$ cm^{-1} is the average splitting of the Kramers doublet of the ground state $^2F_{5/2}$ in the exchange field[11]. We plotted in Fig.3 the temperature variation of the reduced MO coefficient and also the temperature dependence of $C_c(T, \lambda)/C_c(T=50, \lambda)$ for both wavelengths calculated by Xu et al.[3] in the 50 - 294 K temperature range. The similarity of the different curves both in magnitude and temperature variation confirms that the electric dipole transitions contribution to FR of Ce^{3+} ions is predominant[3], [6].

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