

Untangling the contributions of cerium and iron to the magnetism of Ce-doped yttrium iron garnet

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The remarkable magnetic properties of yttrium iron garnets (YIGs) underpin the use of these materials in a broad scope of spintronic and photonic applications. In particular, the addition of rare earth metals in the structure enhances to a great extent the magneto-optical activity, which is beneficial for the development of nonreciprocal optical devices. Exploiting the wavelength selectivity of magneto-optics we have identified a range of frequencies at which one can unravel the individual contributions to the magnetism and gyrotropic response arising from cerium and iron. We envision that this outcome may pave the way to further experiments to assess quantitatively the effect on the optical properties of rare earth incorporation into YIG.

The insulating ferrimagnet yttrium iron garnet (YIG) has come under the limelight over the recent years because of its remarkable properties, taking center stage for a wide range of applications. For instance, the remarkably small damping parameters for spin waves have grabbed the interest for the applications of YIG in magnonics¹, spintronics^{2,3} and spin caloritronics^{4,5,6}. This has been fertile ground for concepts that, for instance, exploit spin waves to pump spins into metals⁷, which can be used to generate thermally driven spin currents^{8,9,10} or magneto-resistance effects induced by spin-orbit coupling^{11,12}. Apart from this, the outstanding magneto-optical properties of YIG have been used in nonreciprocal devices in optical communications^{13,14,15,16,17,18,19}. To further increase its gyrotropic response, YIGs have been doped with rare earths and, in particular, with cerium^{20,21,22,23}. In view of this, untangling the individual contributions of Ce and Fe may provide further clues to a better understanding of the nature of the magneto-optical enhancement in doped YIG and the character of the assigned electronic transitions^{24,25}. In this sense, exploiting the wavelength selectivity of YIG's gyrotropic response may also help solving some controversial issues that are still today a matter of debate, regarding the fundamental mechanisms that rule the magneto-optical enhancement of these compounds²⁶.

For the deposition of Ce-YIG and YIG thin films we used (001)- and (111) oriented gadolinium gallium garnet (GGG) crystals with a thickness of $\sim 500 \mu\text{m}$, which are frequently used for the growth of high-quality YIG films^{27,28}. The samples were grown in oxygen atmosphere by pulsed laser deposition using an excimer laser at 10Hz. Ce-YIG films of composition $\text{CeY}_2\text{Fe}_5\text{O}_{12}$ and thicknesses $t \approx 54 \text{ nm}$ (for (001)-oriented Ce-YIG) and $t \approx 56 \text{ nm}$ ((111)-oriented Ce-YIG) were grown at 880°C in an oxygen partial pressure of 0.3 mbar, whereas (111)-oriented YIG films of composition $\text{Y}_3\text{Fe}_5\text{O}_{12}$ and thickness $t \approx 200 \text{ nm}$ were deposited at 550°C and 0.025 mbar. The Kerr rotation θ and ellipticity ε were measured in a polar Kerr configuration at room temperature for wavelengths in the visible ($\lambda = 400 - 700 \text{ nm}$). Details of the magneto-optical method and equipment can be found elsewhere^{29,30}. Spectra of $\theta(\lambda)$ and $\varepsilon(\lambda)$ were recorded at room temperature by sweeping the wavelengths over the range of the visible in steps of $\Delta\lambda = 1 \text{ nm}$.

First of all, we measured the Kerr rotation and ellipticity of (001)- and (111)- oriented GGG crystals of same nominal thickness ($\approx 500 \mu\text{m}$). The $\theta(\lambda)$ and $\varepsilon(\lambda)$ spectra for both orientations measured at a magnetic field $H \approx 17.5 \text{ kOe}$ turned out to exhibit similar wavelength dependence (Figures 1a and 1b). While Kerr rotation preserved the sign for all wavelengths, the ellipticity reversed sign twice, with two particular frequencies at which the ellipticity vanished, namely, $\lambda_1 \approx 450 \text{ nm}$ and $\lambda_2 \approx 550 \text{ nm}$. This is readily seen in ellipticity loops measured at different wavelengths around λ_1 (Figure 1c). At wavelengths below λ_1 the ellipticity exhibits a linear dependence with field with a positive slope. In contrast, at wavelengths above λ_1 the slope reverses sign (Figure 1c), indicating that the ellipticity decreases on increasing the field. Just in the vicinity of λ_1 the ellipticity is vanishingly small for any value of the magnetic field. The same figure shows the ellipticity data of GGG corresponding to wavelengths around λ_2 .

The selection of wavelengths λ_1 and λ_2 at which the ellipticity of GGG vanishes has been used as the sweet spot to probe the properties of YIG and Ce-YIG films, as it allows to quench the

paramagnetic contribution from GGG. For instance, Figures 2a and 2b display, respectively, the ellipticity of the (001) oriented Ce-YIG film around λ_1 and λ_2 . In both cases, we observed loops that exhibited a positive or negative slope at the highest fields, revealing the contribution of the GGG crystal at wavelengths shorter or longer than λ_1 or λ_2 . Interestingly, Figures 2a and 2b show that for wavelengths in the vicinity of λ_1 and λ_2 the slope of the hysteresis loops at high fields is the smallest; since these are precisely the wavelengths at which the substrate contribution vanishes, we can therefore infer that the ellipticity intrinsic to the Ce-YIG films nearly saturates. Additionally, the hysteresis curves shown in Figure 2c also indicate that the magnetization increases faster along the $\langle 111 \rangle$ direction, in agreement with previous studies reporting on the magnetic anisotropy of YIG³¹.

The previous discussion gives unambiguous evidence that the measurement of ellipticity enables the access to the intrinsic magnetic properties of YIG and Ce-YIG films grown on GGG. We have taken this vantage point to scrutinize their gyrotropic response, with emphasis on untangling any eventual individual contribution from cerium and iron to the magneto-optical activity. A useful insight is provided by the analysis of the spectral dependence of the shape of the hysteresis loops. In the case analyzed here, the ε -loops of Ce-YIG films display an unusual shape when measured at wavelengths in the vicinity of $\lambda = 440$ nm, exhibiting a wavy magnetic field dependence in the central part of the loops (Figures 3a and 3b). As seen in the insets of Figures 3a and 3b, this atypical behavior disappears gradually as soon as the wavelength moves away from $\lambda = 440$ nm, at which point the loops recover progressively the usual shape.

We address now the origin of such anomalous loops. Extracted from SQUID measurements (not shown), the magnetization of Ce-YIG films analyzed in this study turned out to be substantially smaller ($75 - 100$ emu/cm³) than values reported elsewhere³² and bulk (~ 140 emu/cm³). We may speculate whether the existence of some defects, particularly antiphase boundaries, which could cause the magnetization drop, could partly explain the abovementioned atypical shape of loops. Yet, the fact that this phenomenon is only observed for an extremely narrow range of wavelengths (see below) is a clear indication that alternative scenarios have to be envisioned to explain the emergence of anomalous loops. In this sense, the most plausible explanation for the unusual hysteresis is that it arises from the superposition of two independent ellipticity loops with opposite signs. The two-contribution scenario goes a long way towards explaining the observed complex loop behavior and was indeed originally proposed by other authors to explain similar anomalies in the Faraday hysteresis loops of bismuth-doped garnets³³.

Different hypothesis can be put forward to explain the emergence of two distinct hysteresis contributions. One first possibility is that Fe at tetrahedral and octahedral coordination sites contribute differently to the ellipticity³³. Another option is that one of the two contributions is linked to the iron sites –whatever in tetrahedral or octahedral coordination– while the other is related to cerium in dodecahedral sites. To discern which scenario is at play, we have analyzed the spectral response of undoped YIG. As revealed from inspection of Figure 3c, the ellipticity loops of YIG display a conventional shape for all wavelengths, including those straddling the wavelengths at which the ellipticity reverses sign. We conclude, therefore, that the atypical loops observed in

doped YIG are related to the distinctive spectral contributions of opposite signs associated to cerium and iron.

To gain insights into this issue we fitted the experimental hysteresis curves to the expression

$$\varepsilon_{fit} = A_1 \operatorname{erf}\left(\frac{H-H_{c1}}{H_{s1}}\right) + A_2 \operatorname{erf}\left(\frac{H-H_{c2}}{H_{s2}}\right) + A_3 H \quad (\text{Equation 1})$$

where H_{c1} , H_{c2} and H_{s1} , H_{s2} are the coercive and saturation fields of the two contributing loops named Hyst1 and Hyst2, respectively, H is the magnetic field and A_1 , A_2 , A_3 are the weights of the different terms to the simulated ellipticity ε_{fit} . Equation 1 uses the fact that hysteresis loops may be approximated by error functions or hyperbolic tangents^{33,34}. This approximation is sufficient because our main purpose is to disentangle quantitatively the two contributions. For our simulations we have chosen to feed the abovementioned parameters into the error function $\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-t^2) dt$. The first two terms of Equation 1 take account of the two hysteresis curves contributing to the ellipticity, while the last term is plausibly related to the residual ellipticity of the GGG substrates when the gyrotropic response is measured for wavelengths slightly off λ_1 or λ_2 , so that the contribution from GGG is not strictly null.

In the analysis, a least-squares method was used to approximate the Hyst1 and Hyst2 curves to the experimental data. For the quality assessment, the correlation coefficient was used, defined as $r^2 = \frac{\sigma_{\varepsilon\lambda}}{\sigma_{\varepsilon\varepsilon} \times \sigma_{\lambda\lambda}}$, where $\sigma_{\varepsilon\lambda} = \sum_i (\varepsilon_i - \bar{\varepsilon})(\lambda_i - \bar{\lambda})$, $\sigma_{\varepsilon\varepsilon} = \sum_i (\varepsilon_i - \bar{\varepsilon})^2$ and $\sigma_{\lambda\lambda} = \sum_i (\lambda_i - \bar{\lambda})^2$ are the sum of squared values for the set of (ε, λ) data points about their respective means. Figure 4a shows the particular case of the ellipticity measured at $\lambda = 440$ nm. Interestingly, we observe that the Hyst1 and Hyst2 loops shown in Figure 4a display different saturation and coercive fields. To evaluate the accuracy of these values we ran a large set of simulations in which particular values of H_{c1} and H_{c2} were imposed in the fittings, while the saturation fields H_{s1} and H_{s2} were left free to adjust. The $(H_{c1}, H_{c2}, H_{s1}, H_{s2})$ parameter dataset was extracted from the fittings with a margin of error $< 5\%$ with a confidence interval of 95%. Figure 4b shows the correlation coefficient r^2 mapped as function of values in the (H_{c1}, H_{c2}) dataset. We see that the highest-quality fittings were obtained for values $H_{c1} \approx 310$ Oe and $H_{c2} \approx 150$ Oe. Outside this region the values of r^2 were significantly smaller (Figure 4b), indicating that solutions other than those mentioned above were unlikely. At values of (H_{c1}, H_{c2}) where r^2 is the highest, the anisotropy fields are $H_{s1} \approx 5850$ Oe and $H_{s2} \approx 2150$ Oe, respectively. In consequence, the analytical study discussed here is consistent with two contributions that have some degree of magnetic decoupling, one of them arising from cerium and the other from the iron sublattices.

It is only within an extremely narrow window of wavelengths that the two contributing loops combine with opposite signs, yielding the anomalous hysteresis shown in Figure 3. Therefore, it is just inside this limited spectral region that one can reliably deconvolute the two contributions from the as-measured signals. This is illustrated by the analysis of the ε - loops in the range $\lambda \approx 438 - 443$ nm, see Figure 5. At $\lambda \approx 438$ nm (Figure 5a), one of the contributing loops –Hyst2– is almost zero; the corresponding hysteresis curve displays a conventional loop, bereft of any

anomalous shape. In contrast, at $\lambda \approx 441$ nm (Figure 5b), Hyst1 and Hyst2 are finite with opposite signs, and their sum shows the anomalous shape. The magnitudes of Hyst1 and Hyst2 are plotted as a function of wavelength in Figure 5c. Within the interval $\lambda \approx 438 - 443$ nm the as-measured ε -curves can be decomposed into the Hyst1 and Hyst2 loops, thus accessing the individual contributions to the magnetic and gyrotropic response of Ce-doped YIG. Outside this spectral region both contributing loops have the same sign and the mathematical decomposition becomes extremely difficult. Therefore, the differential spectral evolution of the gyrotropic response of each cation sublattice can be used as the optimal condition to access each individual contribution to the magnetism of doped YIG.

We discuss next the most likely assignment of Hyst1 and Hyst2 to the Ce and Fe cations. As mentioned above, the cerium and iron contributions exhibit a certain degree of magnetic decoupling. This observation is consistent with the magnetic behavior of rare earth cations in different types of structures. For instance, in rare earth manganite perovskites, Tb or Dy cations order magnetically at temperatures substantially lower than those of the Mn subnetwork^{35,36}. In the case of garnets, it is generally accepted that the coupling between rare earth cations is weak, and the rare earth-iron exchange interaction barely affects the iron sublattice³⁷. Supporting this view is the characterization of the magnetic susceptibility of Ce-doped fully compensated garnets, in which the paramagnetic contribution can be attributed to the magnetic susceptibility of free Ce³⁺ ions³⁸. In view of all these considerations, the different saturation fields of Ce and Fe may be explained qualitatively by considering that the Fe ions at the tetrahedral and octahedral sites are tightly exchange-coupled and create a molecular field on the magnetic moments at the Ce³⁺ sites. In this scenario, the Ce³⁺ ions, which are weakly coupled to the Fe sublattices, feel the molecular field created by the iron ions and align their moments along this field. Yet, most plausibly, the magnetocrystalline anisotropy arising from the interaction of spins with the lattice has to be considered too. Indeed, spin-orbit coupling is expected to be much larger in *f*- than in *d*-orbitals and, consequently, the contribution of rare earths to the magnetic anisotropy is considerably larger than that of transition metals^{39,40,41}. Therefore, we claim that the magnetization of Ce ions is conditioned by the local magnetic anisotropy at dodecahedral sites, which can be significantly different from that at the Fe sites. This fact would explain why Ce³⁺ magnetic moments may saturate at higher fields than Fe moments. In view of this, it is sensible to assign Hyst1 and Hyst2 (Figure 4a) to the contributions of Ce and Fe, respectively.

In summary, we have identified a narrow spectral range within which the gyrotropic response of Ce-doped YIG can be broken down into two different contributions coming from Ce and Fe. This may also occur for other rare earth doping, possibly for a different frequency range, opening up opportunities to study the properties of doped YIG. For instance, peering into the different sublattice gyrotropic responses may shed light on the specific dynamics of the magnetic moments of the different cations, something that is unattainable for other experimental methods. On the other hand, time-resolved experiments may also provide indirect ways to peer into the nature of the involved transitions, an issue that is still nowadays a matter of debate²⁶. Two basic transitions are proposed: either between 4f and 5d states of Ce³⁺ or between 4f and 3d states of Ce³⁺ and Fe

at the tetrahedral site^{26,42}. Since our experiments –done in quasi-static conditions– show that the magnetization of Ce moments is slightly decoupled from that of Fe, it is expected that dynamic responses can be downright divergent at very short scales –typically below the picosecond where magnetization precession occurs–. In such a scenario, intrasite $Ce^{3+} - Ce^{3+}$ and intersite $Ce^{3+} - Fe$ transitions should yield distinctive optical signatures in such kind of ultrafast experiments. Therefore, ultrafast time-resolved spectroscopy measured at selected wavelengths may offer invaluable indirect information to understand the origin of the optical responses in Ce-doped YIG.

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Figure 1. Spectra of the Kerr rotation $\theta(\lambda)$ and ellipticity (λ) of a (001)-oriented GGG crystal (a) and (111)-oriented GGG crystal (b), measured at a field $H \approx 17.5$ kOe. (c) The ellipticity of the (111)-oriented GGG crystal measured at different wavelengths is plotted as a function of the magnetic field. The wavelengths λ_1 and λ_2 at which the ellipticity of GGG vanishes are identified from this data.

Figure 2. Ellipticity loops of the (001)-oriented Ce-YIG measured in the vicinity of (a) $\lambda_1 \approx 450$ nm and of (b) $\lambda_2 \approx 550$ nm. (c) Ellipticity curves of the (001)- and (111)- oriented Ce-YIG as well as undoped (111)-oriented YIG, measured at $\lambda_1 \approx 450$ nm. The inset shows a zoomed in region of the loops around the highest applied fields.

Figure 3. Panels (a) and (b) show the ellipticity loops of (001)- and (111)-oriented Ce-YIG films, respectively. The displayed curves were measured at wavelengths for which they exhibited an anomalous dependence on the magnetic field. These critical wavelengths were close to the point where the ellipticity curves reverse sign (see the insets). In contrast, undoped (111)-oriented YIG did not show any atypical hysteresis curve at any wavelength, see panel (c). We hence conclude that the anomalous loop shape comes from differential contributions of Ce and Fe to the gyrotropic response.

Figure 4. (a) The ellipticity loop of the (001)-oriented Ce-YIG measured at $\lambda_1 \approx 450$ nm (purple) is broken down into sub-loop components Hyst1 (blue) and Hyst2 (red). For that purpose, fittings to Equation 1 were used. The plot also includes a residual linear dependence on magnetic field (green). (b) The correlation coefficient r^2 of the least-squares fitting is mapped against the values of the coercive fields H_{c_1} and H_{c_2} of the simulated Hyst1 and Hyst2 sub-loops. The points at which r^2 is maximum are indicated by small circles in this chart. (c) and (d) map the values of the saturation fields H_{s_1} and H_{s_2} obtained from fittings to Equation 1; each point of these maps was calculated after fixing in the simulations the values of the coercive fields H_{c_1} and H_{c_2} while H_{s_1} and H_{s_2} were left free to adjust. Small circles in (c) and (d) are at the same location as the maximum value r^2 in (b).

Figure 5. Plots of the as-measured ellipticity loops of the (111)-oriented Ce-YIG film, measured at (a) $\lambda \approx 438$ nm and (b) $\lambda \approx 441$ nm, respectively. The figures include the simulated Hyst1 and Hyst2 loops. The magnitudes of Hyst1 and Hyst2 are plotted in (c) as a function of wavelength within the range $\lambda \approx 438 - 443$ nm. Within this narrow spectral range, the Hyst1 and Hyst2 loops have opposite sign, yielding the anomalous loop shapes displayed in panel (b) and Figure 3. The atypical complex dependence of the ellipticity measured in the range $\lambda \approx 438 - 443$ nm allows a reliable decomposition of the as-measured data into the contributing Hyst1 and Hyst2 loops, enabling the access to the magnetic and optical properties of the Ce and Fe ions.

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