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Large magnetorefractive effect in magnetite

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Abstract. We carried out a magneto-optical spectroscopic study of magnetite (Fe_3O_4) thin films and single crystals at optical wavelengths. We observed a relevant quadratic magneto-optic contribution that can be notably larger than first-order linear magneto-optics at some range of wavelengths in the visible region. These unusual quadratic effects are particularly strong at the Verwey temperature (100–120 K) and decay slowly away from this transition temperature. We attribute this remarkable magneto-optical response to a magnetorefractive effect associated with the field-dependent polaronic conductivity at optical frequencies, which interestingly enough is already noticeable at room temperature.

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1. Introduction

The state of the polarization of light is changed after interacting with a magnetic material, and this phenomenon is exploited in magneto-optical experiments to extract information on magnetic properties [1, 2]. Commonly, first-order linear magneto-optical effects, which are proportional to the components of magnetization, are measured. Depending on the optical arrangement, the longitudinal/polar rotation and ellipticity, which are proportional to the magnetization components in the plane of incidence, or the magneto-optical transverse response, which is sensitive to the in-plane component of the magnetization transverse to the plane of incidence, are detected [1, 2]. In addition, quadratic magneto-optical (QMO) effects, proportional to quadratic products of the magnetization components, are observed in some specific optical arrangements, e.g. in the Cotton–Mouton configuration where the propagation of light is perpendicular to the magnetization [3]. In a general magneto-optical configuration, these quadratic effects, when observed, are usually found to be smaller than the linear contribution, or comparable at most, in some specific materials [3]–[8].

We have recently demonstrated that the magneto-optical response in (La, Pr)CaMnO₃ manganite films is quite complex, with two distinct even-parity contributions that can be unexpectedly large, up to an order of magnitude larger than the linear magneto-optical effects [9]. We attributed one of them to QMO effects increasing with decreasing temperature below the Curie temperature (T_C), with a temperature dependence that mimics that of magnetization [9]. On the other hand, in (La, Pr)CaMnO₃ [9] and other manganites (La_{2/3}Ca_{1/3}MnO₃ and La_{2/3}Sr_{1/3}MnO₃) [10], around T_C an additional contribution was ascribed to a magnetorefractive effect (MRE), i.e. to changes of optical reflectance induced by the applied fields [11, 12]. In that case, the MRE was assigned to a field-dependent polaronic conductivity closely related to the colossal magnetoresistance of ferromagnetic manganites [10]. This unusually complex magneto-optical response should not be restricted to a specific material and, in particular, a significant MRE at visible wavelengths might also be expected close to other metal–insulator transitions where magneto-polaron transport is relevant. Here we show the validity of this assumption by examining the magneto-optical response of magnetite close to the Verwey transition. As in colossal magnetoresistance, hopping polaron conductivity has also been postulated as one of the main mechanisms driving the Verwey transition in magnetite [13, 14]. Confirming our expectations, here we show that magnetite also develops a large magneto-optical response related to the MRE, which is readily observed in a wide range of temperatures up to at least 300 K, being particularly strong at the Verwey temperature.

This indicates that a strong MRE at visible wavelengths is a consequence of the magnetic-field dependence of polaron transport, and that magneto-optics is an invaluable experimental tool to investigate electric transport in strongly correlated systems.

2. Experimental details

In our paper, we analyzed the magneto-optical response of both a magnetite thin film and a single crystal. A magnetite film with thickness $t = 120$ nm was grown on a MgO(001) single-crystal substrate using oxygen-plasma-assisted molecular-beam epitaxy (DCA MBE M600). The growth of the Fe₃O₄ film was carried out by means of electron-beam evaporation of metallic Fe (99.999%) in the presence of free oxygen radicals (1.1×10^{-5} Torr). Details of the growth procedure can be found elsewhere [14, 15]. The Verwey transition occurs at $T_V = 102$ K in this thin film. Additionally, a high-quality Fe₃O₄(001) single crystal was prepared using the skull-melting technique [16]. We measured a Verwey temperature of $T_V \approx 121$ K in the single crystal.

Magneto-optical spectroscopy was carried out in the wavelength range $\lambda = 800$ – 350 nm [10]. Measurements were performed in a range of temperatures $T = 8$ – 300 K and at applied fields up to $H \approx \pm 6$ kOe. These experiments were carried out in a transverse optical configuration, i.e. the magnetic field was applied in the film plane and perpendicular to the plane of incidence, with the light beam incidence set to $\theta = 45^\circ$. The reason for choosing this particular arrangement is that we have sensitivity not only to both rotation and ellipticity, but also to changes of optical reflectance, and thus to MRE [1, 2]. The light source was a Xe 75 W lamp, in combination with a spectrometer. Magneto-optical spectroscopy was based on a PSCA null ellipsometer arrangement [17], in which light coming from the source first goes through a polarizer (P) with azimuth at $+45^\circ$ from the incidence plane (all angles defined positive clockwise looking along wave propagation) before impinging on the sample (S). The polarization state after reflection on the sample is analyzed by the combination of a compensator whose azimuth is set at angles $C = \pm 45^\circ$, and an analyzer (A) before reaching the detector (photomultiplier tube). Between the polarizer and the sample, the light goes through a photo-elastic modulator (PEM), which modulates light polarization. A lock-in amplifier locked to the PEM resonance frequency ($\omega_P = 50$ kHz) is used to measure the signal fed from the light detector at the first harmonic (I_{ω_P}). Our experimental setup allows measuring of a magneto-optical response given by [17]

$$\vartheta[H] = \frac{I_{\omega_P}}{(2I_0 J_1(\varphi_A) \sin 2\Psi)}. \quad (1)$$

In this expression, the magneto-optical signal is an admixture of the imaginary part of the Kerr transverse signal $\Im[\tau_K(H)]$, and the s- and p-components of the magneto-optical rotation and ellipticity $\theta_{s,p}$, $\varepsilon_{s,p}$, respectively, i.e.

$$\vartheta[H] = [(\pm)_C (\pm)_A \Im[\tau_K(H)] + [(\theta_s - \theta_p) \sin \Delta - (\varepsilon_s + \varepsilon_p) \cos \Delta]], \quad (2)$$

where $(\pm)_C$ and $(\pm)_A$ are signs that depend on the ellipsometric null zones for each particular compensator and analyzer azimuths, and (Δ, Ψ) are the ellipsometric angles at zero field [18]. In equation (1), J_1 is the Bessel function ($n = 1$) of the first kind and φ_A is the PEM phase modulation amplitude [18].

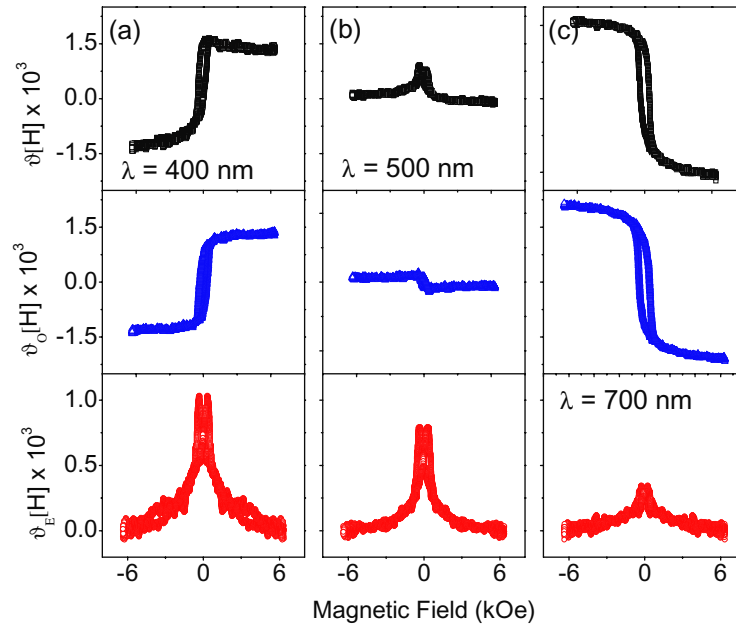


Figure 1. Magneto-optical hysteresis $\vartheta[H]$ loops measured in a magnetite thin film at wavelengths $\lambda = 400$ (a), 500 (b) and 700 nm (c) and at temperature $T = 100$ K. The even ($\vartheta_E[H]$)- and odd ($\vartheta_O[H]$)-parity contributions are also plotted.

3. Results: analysis of the anomalous magneto-optical response

In figure 1, we show the $\vartheta[H]$ data measured in the thin film at wavelengths $\lambda = 400$ nm (figure 1(a)), 500 nm (figure 1(b)) and 700 nm (figure 1(c)) and at a temperature $T = 100$ K. We observe that the hysteresis loops reverse the sign between $\lambda = 400$ and $\lambda = 700$ nm, in agreement with the observed sign reversal of the magneto-optical constants of magnetite in the visible region [19, 20]. More importantly, we see that the shape of $\vartheta[H]$ is strongly dependent on light frequency. For instance, the signal measured at $\lambda = 700$ nm is essentially a symmetric magneto-optical hysteretic loop, i.e. it corresponds to first-order magneto-optical effects, exhibiting odd parity under field inversion $\vartheta[H] = -\vartheta[-H]$. However, at some frequencies, contributions to $\vartheta[H]$ different from first-order conventional magneto-optical effects appear, thus making the shape of $\vartheta[H]$ more complex. These effects are particularly apparent in the data collected at $\lambda = 400$ and 500 nm, as shown in figure 1.

The anomalous shape of the hysteresis loops shown in figure 1 can be analyzed by noticing that the measured signal can be deconvoluted into odd-parity and even-parity components as $\vartheta_O[H] = (\vartheta[H] - \vartheta[-H])/2$ and $\vartheta_E[H] = (\vartheta[H] + \vartheta[-H])/2$, respectively, which are also shown in figure 1. Such decomposition makes it evident that an even-parity $\vartheta_E[H]$ contribution to the magneto-optical signal is particularly strong in some range of frequencies and largely distorts the measured loops. We observed similarly large symmetric $\vartheta_E[H]$ loops in (La, Pr)CaMnO₃ and La_{2/3}(Sr, Ca)_{1/3}MnO₃ thin films [9, 10]. Although not discussed here, we note that the strong wavelength dependence of the even-parity response $\vartheta_E[H]$ observed in figure 1 is related to the change with the magnetic field of the rates of the interband transitions

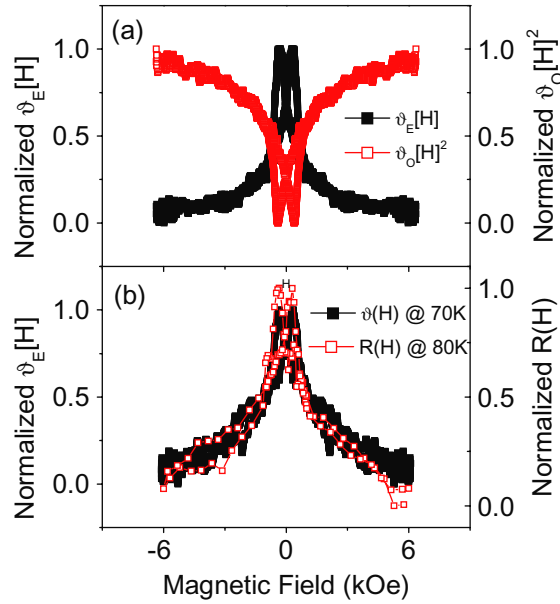


Figure 2. (a) Plot of the normalized values of $\vartheta_E[H]$ ($\lambda = 500$ nm) and $\vartheta_O[H]^2$ ($\lambda = 700$ nm), both measured in the thin film at $T = 100$ K, showing the correlation between $\vartheta_E[H]$ and $\vartheta_O[H]^2$. (b) The normalized $\vartheta_E[H]$ loop measured at $T = 70$ K is plotted together with the normalized values of the magnetic field dependence of the resistance $R(H)$ of the thin film measured at $T = 80$ K.

induced by photons with energies in the visible spectrum [19]. Now we address the origin of such unusual magneto-optical response and identify whether QMO or MRE is responsible for such phenomena.

First, we note that the $\vartheta_E[H]$ and $\vartheta_O[H]$ signals saturate at roughly the same field (see figure 1), thus suggesting that both contributions are correlated. To better illustrate this correlation, we have plotted in figure 2(a), for the thin film, the normalized values of the $\vartheta_E[H]$ loops measured at $\lambda = 500$ nm (figure 1(b)) and the $\vartheta_O[H]^2$ loops measured at $\lambda = 700$ nm (figure 1(c))⁴. The resulting curves are those expected for a $\vartheta_E[H] \propto \vartheta_O[H]^2$ relationship, i.e. the even-parity contributions to the magneto-optical signal are quadratic in the magnetization (M) ($M \sim \vartheta_O[H]$), as expected for QMO effects, and thus at first sight it would indicate that this is their origin. However, as the magnetoresistance in several systems is found to be proportional to $\sim M^2$ [21, 22] and it is known that $MRE \sim MR$, this still leaves the MRE as a possible source of $\vartheta_E[H]$. To check this possibility, we plotted (see figure 2(b)) the $\vartheta_E[H]$ loop measured in the thin film at $T = 70$ K, together with the normalized dc magnetoresistance measured at $T = 80$ K. Figure 2(b) clearly indicates that the observed even-parity $\vartheta_E[H]$ loops measured at optical frequencies are proportional to the hysteretic resistance loops measured in dc transport. This striking coincidence shows that the dc-magnetoresistive response is effectively quadratic in the magnetization, and the even-parity magneto-optical response mimics the field dependence of dc electrical resistivity.

⁴ The $\vartheta_E[H]$ and $\vartheta_O[H]^2$ responses can be compared for any wavelength, reaching the same conclusions. Here, we use $\vartheta_O[H]^2$ at $\lambda = 700$ nm for the sake of clarity, because for this particular wavelength $\vartheta[H]$ is essentially free of the even-parity component.

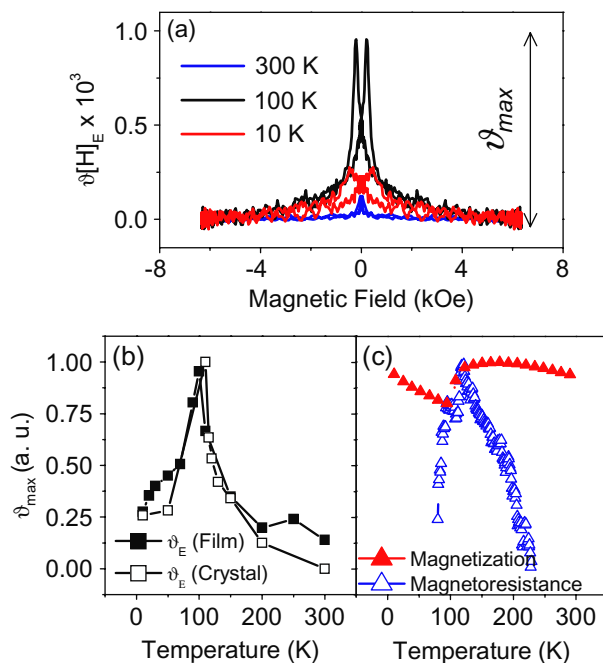


Figure 3. (a) Plot of the normalized even-parity components $\vartheta_E[H]$ of the magneto-optical hysteresis loops of a magnetite thin film measured at $\lambda = 500$ nm and different temperatures. The maximum amplitude ϑ_{\max} of the $\vartheta_E[H]$ signal measured at 100 K is indicated as an illustration. Temperature dependences of (b) ϑ_{\max} measured in both the magnetite single crystal ($\lambda = 600$ nm) and the thin film ($\lambda = 500$ nm) and (c) the normalized values of the temperature dependence of the magnetoresistance $MR(H) = [R(0) - R(70 \text{ kOe})]/R(0)$ (open symbols) and the magnetization (solid symbols) are also plotted. The field was applied in the plane for both cases, and the magnetization was measured at zero field after field cooling at $H = 7$ kOe.

To clarify further the origin of the $\vartheta_E[H]$ loops, we analyzed their temperature dependence. The evolution of $\vartheta_E[H]$ with temperature is illustrated in figure 3(a), where we plot the $\vartheta_E[H]$ loops of the thin film (measured at $\lambda = 500$ nm) at three different temperatures. It is clear from this figure that close to the Verwey transition ($T_V \approx 102$ K), the amplitude of the even-parity contribution is large, i.e. $\vartheta_E[H] \approx 10^{-3}$ at $T = 100$ K, whereas for temperatures away from the transition temperature the quadratic effects, although still clearly visible, are significantly reduced to $\vartheta_E[H] \approx 0.27 \times 10^{-3}$ at $T = 10$ K, and $\vartheta_E[H] \approx 0.12 \times 10^{-3}$ at $T = 300$ K. Similarly, the even-parity contribution measured in the single crystal is also strongest close to the Verwey temperature ($T_V \approx 121$ K in the single crystal) and decays away from this transition temperature: see below and figure 3(b). To illustrate the temperature dependence of $\vartheta_E[H]$ we plotted ϑ_{\max} , defined as the maximum amplitude of the quadratic effect. In figure 3(a), we indicate graphically how ϑ_{\max} is determined for the $\vartheta_E[H]$ loop measured at $T = 100$ K. In figure 3(b), we show the values of ϑ_{\max} measured for both the thin film and the single crystal at different temperatures in the range of $10 \text{ K} \leq T \leq 300 \text{ K}$. This figure clearly shows that the even-parity contributions are in both cases strongly peaked at the respective Verwey temperatures—notice the small shift to the lower temperature of the peak of the thin film with

respect to that of the single crystal, in agreement with the higher Verwey temperature of the single crystal. Note also the relatively slow decay with temperature away from the transition, making the effect observable even at room temperature, i.e. about 200 K above the transition.

The close agreement of the temperature evolution of ϑ_{\max} and the Verwey transition suggests that the anomalous magneto-optical response is correlated to the electrical transport properties of magnetite; see e.g. [23, 24] for extensive reviews on this topic. To check this correlation, we plotted in figure 3(c) (open triangles) the temperature dependence of the normalized magnetoresistance measured in the thin film, defined as $\text{MR}(H) = [R(H=0) - R(H=70 \text{ kOe})]/R(H=0)$. We observe that, similarly to the amplitude of the even-parity $\vartheta_E[H]$ magneto-optical response, the $\text{MR}(H)$ curve is also peaked at the Verwey temperature (figure 3(b)), in agreement with a strongest MR close to the transition reported previously [14, 25]. In contrast, the temperature dependence of the magnetization (figure 3(c), solid triangles) differs strongly from that of both $\vartheta_E[H]$ and MR. Both the magnetoresistance and the magnetization displayed in figure 3(c) were measured in the thin film by applying the field along a [001]-direction in the film plane, and the magnetization was measured in zero field after cooling the sample under a field $H = 7 \text{ kOe}$. The data shown in figure 3(c) further support the correlation of $\vartheta_E[H]$ with MRE. One notices that the temperature dependence of the quadratic magneto-optical response, having a maximum at $T \approx T_V$ as shown in figure 3(b), mimics the temperature dependence of the diffuse neutron scattering intensity observed in magnetite crystals [26], which has been taken as a signature of the presence of uncorrelated polarons that survive well above the Verwey transition [13, 27]. Our observation suggests that the MRE response observed in these magneto-optical experiments arises from magnetic-field-dependent polaronic transport. Interestingly enough, the magneto-polaronic effects extend well above T_V , up to room temperature.

4. Summary and conclusions

To summarize, we have carried out magneto-optical spectroscopy in the visible region for both the magnetite thin film and the single crystal. We show that the hysteresis loops can be dramatically distorted to exhibit a conspicuous asymmetry that can be explained in terms of an unusually large quadratic contribution to the first-order linear magneto-optical coefficients. The effect is also dependent on temperature, and in particular is strongest at the Verwey temperature. We attribute such quadratic effects to a magnetorefractive effect, resulting from the changes of optical conductivity with field through the Verwey transition. The observation of this phenomenon in both a thin film and a single crystal shows that the effect presented here is inherent to the intrinsic electronic properties of magnetite, and confirms that an unusually large contribution from nonlinear magneto-optics terms can be observed in different magnetic materials, for an appropriate range of frequencies.

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