Strain-magneto-optics in ferrite-spinel – new magneto-optical phenomena associated with magnetoelastic interactions

A V Telegin*, Yu P Sukhorukov and N G Bebenin
M.N. Mikheev Institute of Metal Physics, UB of RAS, 620137 Ekaterinburg, 18 S Kovalevskoi Street, Russia
*telegin@imp.uran.ru

Abstract. The correlation between magnetoreflection, magnetotransmission of unpolarized light in the infrared range and the magnetoelastic properties of magnetics is reported for the ferrimagnetic spinel \( \text{CoFe}_2\text{O}_4 \) single crystal. The new physical mechanism responsible for the spectral and field-dependent peculiarities of magnetoreflection and magnetotransmission in the ferrite-spinel possessing strong magnetostriction is discussed. The influence of a magnetic field on specular reflection and transmission spectrum is likely to be indirect: magnetic-field-induced strong strains and deformations of the crystal lattice lead to the change in the electron energy structure and, hence, optical properties of \( \text{CoFe}_2\text{O}_4 \). The revealed mechanism of new infrared magnetooptical effect in magnetostrictive magnetics paves the way towards new research area called as a strain-magneto-optics.

1. Introduction

Giant magneto-optical (MO) effects in magnetic semiconductors connected with the influence of a magnetic field on absorption and reflection of light have been intensively studied starting from the last century due to the industrial demands [1-3]. Some intriguing MO phenomena for unpolarized light have been found in ferromagnetic manganites and spinel in the infrared (IR) range [4]. These effects originally were described as magnetic-field induced changes of light reflection – magneto-reflection effect:

\[ \Delta R/R = (R_H - R_0)/R_0, \]  

refractive index – magnetorefractive effect:

\[ \Delta n/n = (n_H - n_0)/n_0 \]  
and transparency – magneto-transmission effect:

\[ \Delta t/t = (t_H - t_0)/t_0, \]  

where \( R_H, n_H \) and \( t_H \) are the specular reflection, refractive index and transmission of light in the presence and absence of magnetic field. The main mechanisms of the giant MO effects in the IR have been clarified in [5]. Almost 30 years ago N.G. Bebenin made a prediction of the possible influence of magnetoelastic deformations on absorption of light in spinel in the near IR spectral range [6]. It seems quite natural to choose a magnetic with the highest magnetostriction value like as \( \text{CoFe}_2\text{O}_4 \) for studying this MO phenomena. However, only recently a new MO effect connected with the influence of magnetoelastic deformations on absorption of IR unpolarized radiation in \( \text{CoFe}_2\text{O}_4 \) spinel has been observed [7,8]. The change in light intensity in the IR range under the field can reach tens of percent.
and substantially exceeds the effects in polarized light. This research has triggered a new branch of straintronics called as infrared strain-magneto-optics and the study of the change of MO properties of magnetics due to mechanical deformations of various origin.

In the manuscript, we presented the optical and MO properties of the ferrimagnetic CoFe$_2$O$_4$, where the direct correlation between magnetoreflection and magnetotransmission of unpolarized IR radiation with magnetostriction of spinel was shown. We also compared the MO phenomena observed in unpolarized light with the Faraday effect and made some estimations for practical application.

2. Experiment

2.1. The benefits of CoFe$_2$O$_4$

The CoFe$_2$O$_4$ ferrite-spinel seems to be the most promising object for IR strain-magneto-optics due to a few reasons. Firstly, it has the highest value of magnetostriction $\Delta l/l$ at room temperature [9] ($\Delta l$ is the extension or shortening of a magnetic under the influence of a magnetic field) compared to other magnetic semiconductors or dielectric materials [10]. Secondly, its Curie temperature ($T_C$=812 K) is high. And finally, its high transparency in the IR range due to absent of free charge carriers (DC resistivity value is $\rho$\approx10$^5$ $\Omega\cdot$cm).

2.2. Material and methods

The single crystals of CoFe$_2$O$_4$ was grown by floating zone melting method. Correspondence of chemical composition to CoFe$_2$O$_4$ per formula unit was confirmed by X-ray microanalysis. Magnetization data were obtained with the help of Lake Shore 7400 vibrating sample magnetometer. The magnetostrictive properties were characterized by strain gauge technique using the (001) oriented plateshaped samples. The optical properties of samples were collected by IR prism monochromator. The specular reflection coefficient was calculated as $R=I_S/I_{Al}$, where $I_S$ and $I_{Al}$ are the intensities of the unpolarized light reflected from a sample and the Al mirror, respectively. The magnetorefractive effect $\Delta n/n$ was calculated by the Kramers-Kronig method using data for the specular reflection coefficients. The absorption coefficient $K$ was determined by the expression

$$K = (1/d) \ln[(1 - R)^2(1/t)],$$

where $d$ is the thickness of the sample.

2.3. Magnetization and magnetostriction

The field dependences of magnetization $M(H)$ has the saturation value of 78 emu/g at coercive force $H_c$=80 Oe and at $T$=295 K (Figure 1a). This value of magnetization is close to that reported for a high-quality single crystal [11]. For angle $\phi = 45^\circ$ between $\mathbf{H}$ and the (100) axis or $\mathbf{H}||$(110) the magnetization curve has two steps which are associated with distortion of cubic symmetry of the crystal [12]. The magnetostriiction data obtained coincide well with the reference ones from [9,13,14]. For example, magnetostriction data obtained in spectrometer (range of crystal length along the [100] axis upon magnetic field) reaches $-221$-10$^{-6}$ for $\mathbf{H}||$(010) and $-624$-10$^{-6}$ for $\mathbf{H}||$(100) at room temperature (Figure 1b and 1c). The angular dependence of magnetostriction for the cubic crystal can be described by equation:

$$\frac{\Delta l}{l} = \frac{3}{2}A_{100} \left[ a_1^2 + a_2^2 + a_3^2 + 2a_1a_2a_3 + \frac{1}{2}a_4^2 \right] + 3a_{111} \left[ a_1^2 + a_2^2 + a_3^2 + a_4^2 \right]$$

where $a_{1,2,3}$ are the directional cosines of magnetization vector and $\beta_{1,2,3}$ are the elongation directional cosines. In our case $a_1 = a_2 = a_3 = 0$ and $\beta_1 = 1$. The magnetostriction reaches maximum at $\mathbf{H}||$(100) and is two times less ($\Delta l/l)_{100} = A_{111}/2$ with opposite sign at $\mathbf{H}||$(010). In the case of $a_1 = 1/\sqrt{3}$ ($\phi=54^\circ$) the magnetostriction is ($\Delta l/l)_{100} = 0$. The crystal deformation along a fourfold axis due to magnetostriction are supposed to lead to significant changes in the electronic structure and changes in the shape of absorption and reflection spectra, respectively.
2.4. General optical features

The well-known fact is the importance of IR optical spectroscopy to study the electronic structure, impurity states and charge carriers in materials. Meanwhile there was a lack of optical data for CoFe$_2$O$_4$ crystal in the IR spectral range. The obtained absorption spectrum of the CoFe$_2$O$_4$ (Figure 2a) is in good agreement with the optical conductivity spectrum (Figure 2c) calculated by the Kramers–Kronig method from the specular reflection (Figure 2b).

![Figure 1](image1.png)

**Figure 1.** Field dependences of (a) magnetization $M$ for $H||[100]$ and $H||[110]$ ($\phi = 45^\circ$); (b, c) magnetostriction $(\Delta l/l)_{100}$ at $T = 295$ K for CoFe$_2$O$_4$.

![Figure 2](image2.png)

**Figure 2.** Spectra of (a) light absorption $K$ of the CoFe$_2$O$_4$ at different temperatures, (b) specular reflection coefficient $R$, and (c) the optical conductivity $\sigma$ at $T=295$ K. On inset – spectra of $K$ in the presence and absence of magnetic field.
The increase of $K$ at wavelength $\lambda < 2 \mu m$ is related to the low-energy fundamental absorption edge at $E_g = 1.18$ eV ($\sim 1 \mu m$). The latter is formed by indirect interband transitions from the hybridized $d_{Co} + p_{O}$ states of the valence band at the X point to the $d_{Fe}$ states of the conduction band at the $\Gamma$ point of the Brillouin zone [15]. On cooling from 300 to 80 K, the absorption edge undergoes a “blue” shift of about 8 meV, which is close to the data reported in [16]. A weak band at $\lambda = 2.6 \mu m$, which can be seen in the optical conductivity spectrum, is related to vacancies in the anion sublattice [17]. Low resolved absorption bands at $\lambda = 6$, 8 and 10 $\mu m$ can be attributed to the impurity levels either in the anion and/or cation sublattice of CoFe$_2$O$_4$ [17]. The observed specular reflection spectrum is formed by the low-energy absorption edge at $\lambda < 2 \mu m$ and the phonon bands at energies $E_1 = 609$ cm$^{-1}$ ($\lambda = 16.4 \mu m$), $E_2 = 413$ cm$^{-1}$ ($\lambda = 24.2 \mu m$), $E(T_{1u}) = 466$ cm$^{-1}$ ($\lambda = 21.5 \mu m$) and $E(T_{2u}) = 534$ cm$^{-1}$ ($\lambda = 18.7 \mu m$) as well as the frequency-independent part of reflectance ($R \sim 14.7 \%$) at 2 $\mu m < \lambda < 7.5 \mu m$. Phonon band $E_1$ is associated with vibrations of Co–O ions in the octahedral sublattice and the $E_2$ band is connected with the vibrations of oxygen in the tetrahedral sublattice [18,19]. The application of the magnetic field lead to changes in absorption (inset in Figure 2a) and reflectivity, i.e. magnetoabsorption (magnetotransmission), magnetoreflection and magnetorefractive effects have appeared.

2.5. General magnetooptical features

In CoFe$_2$O$_4$ we observed large values of MO effects for unpolarized light in the IR range (Figure 3). The general features of magnetooptical effects in the spinel crystal are closely connected with aforementioned optical features of absorption and reflection spectra of the spinel (Figure 2).

![Figure 3. Spectra of (a) magnetoreflection $\Delta R/R$, (b) magnetorefractive effect $\Delta n/n$, and (c) magnetotransmission $\Delta t/t$ at $T=295$ K and $H=3.6$ kOe in the Faraday (1) and Voight (2) experimental geometry at different direction of magnetic fields relative to crystallographic axes of CoFe$_2$O$_4$ crystal.](image)

Connection with the optical features are manifested (i) in the increase of MO effects at $\lambda < 2 \mu m$ as a result of the shift of the absorption edge; (ii) in the appearance of spectral peculiarities associated with the change of the intensity and peak position of MIR bands at $\lambda = 2.96 \mu m$, 6 $\mu m$ and 8 $\mu m$, and (iii) the shift of the reflectance minima in the vicinity of phonon bands at $\lambda > 9 \mu m$ under the action of the magnetic field [12,17]. The evaluation of the magnetoreflection by the expression
\[ \Delta R/R = [4(n^2-k^2-1)\cdot \Delta n + 8n'k'\cdot \Delta k]/[(n+1)^2 + k^2] \],

(6)

developed in [20] for the case of normal incidence of light and in the absence of free charge carriers, gives a good agreement with the experiment data by exception the features associated with the displacement of the reflection bands. Thus, one can apply the theory of magnetoreflection and magnetorefractive effects for description MO effects in spinel in the IR range.

Other indications of correlation of magnetoreflection and magnetotransmission effects with magnetostriction are manifested (i) in their dependences on the orientations of the crystal in the magnetic field similar to magnetostriction, (ii) in the deformation-induced displacement of the top of the valence band, (iii) in the similarity of the magnetic field dependences of the effects in the Faraday and Voight geometry of experiment [12,17]. We supposed the effect of the applied magnetic field on the light absorption is indirect: the magnetic field causes the distortion of the crystal lattice and lead to significant changes in the electronic structure and varying the shape and position of absorption and reflection spectra. The estimation of deformation potential for the valence band gives value about 20 eV, which is consistent with the available data on the band ferrite structure and has a value close to the experimental data near the absorption edge. The calculations give about 0.5% for \( \Delta R(\lambda)/R \) and ~1% for \( \Delta t(\lambda)/t \) in Voight geometry [6]. The difference in magnitude of \( \Delta t/t \) in the Voight and Faraday geometry (curves 1 and 2 in Figure 3c) can be explained by the additional contribution of mechanical deformation stresses, which reaches their maximum when the field is applied perpendicular to the sample surface.

In addition, we selected the spectral region 4 µm<λ<12 µm in which the magnetorefractive effect \( \Delta n/n \) exists (Figure 3b). The value of \( \Delta n/n \) was found to be about \( +1.5 \cdot 10^{-3} \) for \( H=3.6 \) kOe and \( T=295 \) K. It is an order of magnitude larger than that predicted for orthoferrite YFeO\(_3\). However, the magnetostriction in YFeO\(_3\) is an order of magnitude less than in CoFe\(_2\)O\(_4\) [21]. Therefore, we can say that magnetorefractive effect in CoFe\(_2\)O\(_4\) is also associated with its magnetoelastic properties.

2.6. Field dependencies

As it has been already mentioned the direct correlation between the effects of \( \Delta R/R \), \( \Delta t/t \) and \( (\Delta l/l)_{100} \) can be seen from their field dependencies (Figure 4 and 5). In contrast to magnetization \( M(H) \) (Figure 1a) \( \Delta l/l(H) \), \( \Delta R/R(H) \) and \( \Delta t/t(H) \) dependencies are even function and are determined by the change of diagonal components of the permittivity tensor (\( \varepsilon_{ii} \)). For \( |H|(100) \) the magnetoreflection and magnetotransmission as well as magnetostriction demonstrate an abrupt increase starting from \( H \approx 1.7 \) kOe, and reach saturation at \( H > 2.5 \) kOe (Figure 4).

The complex absorption spectra of spinel crystal and its high sensitivity to external magnetic field leads to a complex behavior of the \( \Delta R/R(\lambda, H) \) and \( \Delta t/t(\lambda, H) \) (Figures 3,4). For example, the change of MIR-bands as well as low-energy slope of the fundamental absorption edge in the magnetic field leads

![Figure 4. Field dependences of (a) magnetostriction \( (\Delta l/l)_{100} \), (b) magnetoreflection \( \Delta R/R \) at \( \lambda=2.7 \) µm and (c) magnetotransmission \( \Delta t/t \) at \( \lambda=3.4 \) µm and at angle \( \phi =40^\circ \) at room temperature for \( |H|(100) \).](image-url)
to an unusual shape of $\Delta R/R(\lambda)$ and $\Delta t/t(\lambda)$ in the IR region (curve $\phi$ in Figure 4c). It is obtained for the first time that the sign and magnitude of the $\Delta R/R(H)$ and $\Delta t/t(H)$ effects in spinel crystal can be controlled by only varying the magnetic field for a fixed wavelength. The different behavior of $\Delta R/R(H)$ and $\Delta t/t(H)$ is in case of $H||$(010) (Figure 5). Just like magnetostriction the $\Delta R/R(H)$ and $\Delta t/t(H)$ appear in weak magnetic fields, then smoothly increase and are saturated in the fields $H \geq 2.5$ kOe as in the case of $H||$(100). Thus, $\Delta R/R(H)$ and $\Delta t/t(H)$ strictly follow the $(\Delta l/l)_{100}$ behavior and their correlation is evidently proofed.

If one denotes the parameters $R_{(100)}$, $t_{(100)}$ and $R_{(110)}$, $t_{(110)}$ as directions of magnetization along the axes (100) and (110), respectively, the dependence of these parameters on the orientation of field $H$ in saturated state can be expressed in the form:

$$R = R_{(100)} + 4(R_{(110)} - R_{(100)}) \alpha_x^2 \alpha_y^2$$  \hspace{1cm} (7)

$$T = t_{(100)} + 4(t_{(110)} - t_{(100)}) \alpha_x^2 \alpha_y^2.$$  \hspace{1cm} (8)

Equations (7) and (8) allow determining the magnitude of the MO effects at different angles $\phi$ between direction of the magnetic field and crystallographic axis. For example, in the angle interval $45^0 < \phi < 55^0$ the value of the magnetotransmission and magnetoreflection must be close to zero, which is coincide with experimental data (Figures 3a, 3c and 5c).

![Figure 5. Field dependences of (a) magnetostriction $(\Delta l/l)_{100}$, (b) magnetoreflection $\Delta R/R$ at $\lambda = 2.7$ μm, and (c) the magnetotransmission $\Delta t/t$ at $\lambda = 3.4$ μm and at angle $\phi = H^\wedge(110) - 58^0$ at room temperature for $H||$(010).](image)

2.7. Faraday rotation

In real experiment some partial polarization of light by the optical system ($P=0.1$ in our case) should be taken into account. The question arises of whether the Faraday rotation ($F$) influences on the observed magnetotransmission effects. The contribution of $F$ to the magnetotransmission in our case can be detected as asymmetry in the field dependences of $\Delta t/t(H)$ during commutation procedure, for example, at $\lambda=2.3$ μm (Figure 6).
Figure 6. Field dependence of magnetotransmission in the Faraday geometry of experiment at room temperature and \( \lambda = 2.3 \ \mu m \).

The sign and value of asymmetry correspond to the spectra of the Faraday effect for CoFe\(_2\)O\(_4\) in the IR range [22]. The estimated from the Malus’s law variation of the intensity because of Faraday rotation in the saturated magnetic fields yields the magnitude almost ten times less than that in the magnetotransmission effect.

3. Conclusion  

The direct correlation between the magnetoreflection and magnetotransmission of natural light in the IR range with magnetostriction of a magnetic was for the single-crystal of CoFe\(_2\)O\(_4\) shown. This correlation is confirmed by the similarity of the field behavior of effects at different orientations of magnetization with respect to crystal axes. The mechanisms of magnetotransmission, magnetoreflection of light and magnetorefractive effect in spinel connected with the magnetic-field-induced mechanical deformation of spinel crystal were revealed. It was also shown that the effect of the magnetic field on the optical properties of spinel is indirect: the magnetic field strongly affects the crystal lattice, which in turn leads to the changes in the absorption and reflection spectra. The contribution of traditional Faraday rotation on the magnetotransmission effect in the IR range is quite small.

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