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Magnetorefractive Effect and Cubic Nonlinear Magneto-optics in Magnetic Granular Alloys

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We present a brief review of recent experimental and theoretical results on new magneto-optical phenomena in magnetic granular metal-insulator alloys with tunnel-type magnetoresistance, focusing on magnetorefractive effect and cubic nonlinear magneto-optics.

Key words : magnetorefractive effect, granular alloys, nonlinear magnetooptics

1. Introduction

Recently, new effects and artificially fabricated micro- and nanostructured materials have been proposed to enhance both linear and nonlinear magneto-optical phenomena in magnetic granular metal-insulator alloys in the visible and infrared region of spectra [1-4].

Magneto-optical spectra of granular alloys have been investigated for nearly three decades, but it is still a subject of intense discussion. Traditional magneto-optical effects are due to the influence of spin-orbit interaction on intraband and interband optical transitions. As a rule, magneto-optical response in granular alloys is of the same order of magnitude as that in bulk alloys, namely about 0.1% at visible wavelengths and less than 0.01% at infrared wavelengths in reflection mode [5, 6]. The observed magneto-optical spectra in most cases can be explained in the framework of the effective medium theory (see [7] and references therein), except the experimental data for MnAs-GaAs alloys [8] and discontinuous multilayers [9], in which magneto-optical response is enhanced by one order of magnitude, but it is still small.

Recently, it has been shown that, when the one-dimensional photonic crystals are composed of dielectric layers with built-in magnetic granular alloy layer (magnetophotonic crystals), they exhibit remarkable magneto-optical properties accompanied by two orders of magnitude enhancement in their Kerr rotation [3]. The unique properties arise from the weak localization of light as a result of multiple interference of light within magnetophotonic crystal [10].

Besides traditional magneto-optical effects connected with spin-orbit interaction, and their enhancement in magnetophotonic crystals, a new magneto-optical effect in nanostructured materials with large magnetoresistance, so- called magnetorefractive effect (MRE) [1, 2], was discovered and studied thoroughly. The effect consists of changes in optical properties of systems with large magnetoresistance when they are magnetized [1, 2, 11-13]. Since optical properties are determined by the dielectric function $\varepsilon(\omega)$, which is linear with frequency-dependent conductivity $\sigma(\omega)$, reflection as well as transmission and absorption of light for any kind of magnetic materials with large magnetoresistance strongly depend on magnetization. The effect exists in the infrared region of spectrum in which the interband transitions and interband optical conductivity do not play a dominating role. MRE was discovered first in multiayers [1, 2], next, in granular alloys Co-Ag [12, 13], and recently, in nanocomposites Co-(Al-O) [14], CoFe-(Al-O) [15], CoFe-MgF [16], and (CoFeZr)-SiO_n [17]. MRE is a frequency analogue of magnetoresistance and therefore, its main mechanism in nanocomposites with tunnel-type magnetoresistance is high frequency spin-dependent tunneling.

At finite frequencies, the nonlinear dependence of electrical displacement $\boldsymbol{D}$ on electric field $\boldsymbol{E}$ is the basis of nonlinear optics and magneto-optics. Nonlinear optical

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and magneto-optical phenomena can be observed in the case of high harmonic generation and at the same frequency of incident light. Magneto-induced second harmonic generation and so-called nonlinear magneto-optical Kerr effect [18, 19] are the well-known examples of the former case. It has been shown that the magnetic contrast of magneto-induced second harmonic generation in magnetic granular alloys can be as high as 0.3 and there is a correlation between magnetic contrast and magnetoresistance [20]. Nonlinear magneto-optics at the frequency of incident light was observed in magnetic garnets [21], and for the best of our knowledge it has not been studied in magnetic granular alloys.

In this paper we are discussing MRE at infrared wavelengths and nonlinear magneto-optics in granular metal-insulator alloys at the frequency of incident light.

2. Magnetorefractive effect in nanocomposites: Theory

The reflectivity R of light in a semi-infinite medium model (air and sample) at normal incidence is

$$R=\frac{(1-n)^2+k^2}{(1+n)^2+k^2}, \tag{1}$$

In Eq. (1) $n^*=n-ik$ and we neglect all effects connected with the influence of spin-orbit interaction on optical constants. Since the complex dielectric function

$$\varepsilon^*=(n^*)^2=\varepsilon'-i\varepsilon''$$

is

$$\varepsilon^*=\varepsilon_r-i\frac{4\pi\sigma(\omega)}{\omega}, \tag{2}$$

and the tunnel junction can be thought of as a capacitor [22], the optical conductivity of granular metal-insulator alloys with composition close to the percolation threshold can be written as

$$\sigma(\omega,H)=\frac{1+\frac{\varepsilon_{ins}}{4\pi}\omega\rho(H)}{\rho(H)}, \tag{3}$$

where $\rho(H)$ is the resistivity and ε_{ins} is the dielectric constant of the insulator between granules. For simplicity in Eq. (3) we consider a flat capacitor and do not average Eq. (3) over granular size distribution and distances between them because of only a few tunnel junctions are responsible for transport close to the percolation threshold. Then it immediately follows from Eqs. (1-3), that MRE defined as

$$\frac{\Delta R(\omega,H)}{R}=\frac{R(\omega,H=0)-R(\omega,H)}{R(\omega,H=0)}, \tag{4}$$

is

$$\frac{\Delta R}{R}=-(1-R)\frac{\Delta\rho}{\rho}k^2\left[\frac{3n^2-k^2-1}{(n^2+k^2)[(1-n)^2+k^2]}\right], \tag{5}$$

where the optical constants correspond to the demagnetized state and magnetoresistance

$$\frac{\Delta\rho}{\rho}=\frac{\rho(H=0)-\rho(H)}{\rho(H=0)}$$

is positive.

According to Eq. (5) the MRE frequency dependence in nanocomposites differs from that for metallic granular alloys [2], MRE can be negative as well as positive and it is large if reflectivity is small.

Using Fresnel formula it is straightforward to generalize the above approach to an arbitrary incident angle and polarization. Besides, it is not difficult to apply the approach to the three-layers model (air- sample-substrate) when MRE can be enhanced due to interference of light.

The Eq. (5) is valid if the tunneling probability does not depend on frequency, which is correct for infrared wavelengths [23]. Besides, we can neglect phonon-assistant tunneling because of low intensity of light in the experiment.

3. Magnetorefractive effect in nanocomposites: Experiment

3.1. Correlation between MRE and magnetoresistance

MRE is an even magneto-optical effect, namely it is linear with magnetization squared, like magneroresistance, and therefore as a first step it is necessary to prove that the observed changes in reflection by applying magnetic field are not due to the traditional even magneto-optical effect [24] which is connected with spin-orbit interaction. In fact, both the diagonal and non-diagonal components of the dielectic function depend on magnetization because of influence of spin-orbit interaction and it leads to the orientational magneto-optical effect [24], which is no larger than 0.01% at visible wavelengths, because of spin-orbit interaction and magneto-optical factor are less than 0.1. Therefore, one can neglect the traditional even magneto-optical effect when MRE is larger than 0.1%. Precise measurements of MRE and magneto-optical effects in (CoFeZr)- SiO_n alloys [17] confirm these considerations. Besides, the direct evidence that the observed changes in reflection are due to MRE is the observed correlation between MRE and magnetoresistance (see Eq. (5)). It has been shown that this correlation takes place for both metal-metal [12, 13] and metal-insulator [16, 17] alloys. The correlation was observed for both field and concentration dependences of MRE and magnetoresistance [16, 17]. For example, MRE in $(CoFeZr)_x(SiO_n)_{1-x}$ is insignificant for compositions far from the percolation threshold and increases with magnetoresistance close to the percolation threshold.

3.2. Correlation between MRE and optical properties

The Eq. (5) predicts the strong dependence of MRE on optical properties. It follows from Eq. (5) that MRE increases when reflection is small and absorption is large.

This conclusion is in a good agreement with the experiment for all the studied granular alloys. For example, MRE in CoFe-(Al-O) alloys was observed only at frequencies corresponding to the strong absorption of light [15], MRE reaches its maximum value in (CoFe)-(MgF) alloys when reflectivity is about 5% (Fig. 1) [16], there is the correlation between MRE and reflection and absorption in $(CoFeZr)$-SiO_n alloys [17]. Perhaps, opposite signs of MRE in $(CoFe)_{15}(Al_2O_3)_{85}$ and $(CoFe)_{25}(Al_2O_3)_{75}$ alloys [15] are due to the change of sign of the factor $3n^2-k^2-1$ in Eq.(5).

However, there are some features that cannot be explained by Eq. (5) in the case of (CoFe)-(MgF) alloys (Fig. 1) [16]. First, MRE is approximately the same for 5.7 and 20 μm in spite of reflectivity 4 times smaller for 5.7 μm. Second, MRE exhibits two-peak resonance behavior around 10 μm with no indications for noticeable changes in reflectivity for this frequency. Third, the large

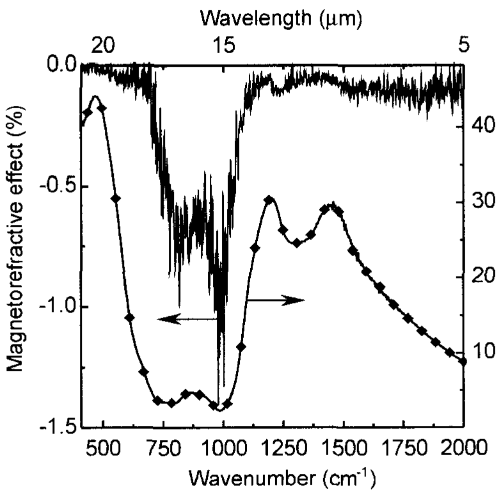

Fig. 1. Magnetorefractive effect at H=1.7 kOe and reflectivity versus light wave number (bottom axis) or wavelength (upper axis) for nanogranular films $(Co_{0.4}Fe_{0.6})_{48}(MgF)_{52}$ at the incident angle $\varphi = 8^0$.

fluctuations in MRE are inherent (CoFe)-(MgF) samples and have never been observed in other systems.

It indicates indirectly the existence of an additional mechanism of MRE in (CoFe)-(MgF) nanogranular films, probably, connected with some excitations in the matrix MgF.

3.3. Angular dependence

Up to now there has not been performed systematic investigations of MRE dependence on incident angle and on polarization of light. Some preliminary measurements [16, 17] showed that MRE reaches maximum value at normal incidence. It is consistent with the Frensnel formula for reflectivity and the approach described above.

3.4. How to increase MRE?

The observed MRE amplitude in granular metal-insulator alloys is between 0.1 and 1.5%. The record value 1.5% has been found in $(Co_{0.4}Fe_{0.6})_{48}(Mg\text{-}F)_{52}$ alloy which exhibit small reflectivity and large magnetoresistance (7.5%) in a relatively weak magnetic field (1.7 kOe). It is at least two orders of magnitude larger than traditional magneto-optical Kerr effects in metallic systems at the same frequencies. The theory predicts a possibility for even larger MRE values by tuning the optical constants.

There are at least two ways to further increase of MRE in nanocomposites. First, MRE is larger in the case of interference that was demonstrated in [14]. Secondly, we can predict that MRE may be extremely enhanced in magnetophotonic crystals based on granular alloys [3] because of multiple reflections and absorption.

4. Cubic nonlinear magnetooptics

Let us consider nonlinear magnetooptics in granular metal-insulator alloys at the frequency of an incident light, which is due to a weakly nonlinear relation between $\boldsymbol{D}$ and $\boldsymbol{E}$ of the form

$$\boldsymbol{D}_i = \varepsilon_i \mathbf{E}_i = \varepsilon_i^{(0)} \mathbf{E}_i + \chi_i^{(3)} |\boldsymbol{E}_i|^2 \boldsymbol{E}_i \,;\; \chi_i^{(3)} |\boldsymbol{E}_i|^2 << \varepsilon_i^{(0)}. \qquad (6)$$

for both constituent materials ($i = 1$ for metal and $i = 2$ for insulator). We assume that linear $\varepsilon_i^{(0)}$ and cubic nonlinear $\chi_i^{(3)}$ dielectric functions have diagonal and linear with magnetization non-diagonal components as follows:

$$\varepsilon_i^{(0)} = \begin{pmatrix} \varepsilon_i^d & -\varepsilon_i^{od} & 0 \\ \varepsilon_i^{od} & \varepsilon_i^d & 0 \\ 0 & 0 & \varepsilon_{zi}^d \end{pmatrix} \quad \chi_i^{(3)} = \begin{pmatrix} \chi_i^d & -\chi_i^{od} & 0 \\ \chi_i^{od} & \chi_i^d & 0 \\ 0 & 0 & \chi_{zi}^d \end{pmatrix}. \qquad (7)$$

Since non-diagonal components of dielectric function are responsible for magneto-optical effects, one can expect that magneto-optical effects in such materials and composites depend on light intensity. It is quite difficult to observe this nonlinear effect for homogeneous ferromagnets because of rather small magnitude of cubic non-diagonal term χ_i^{od}. Using the effective medium approximation (EMA) we show below that magneto-optical cubic nonlinearity in nanocomposites can be at least three orders of magnitude greater than that for constituent materials. Moreover, a composite may exhibit nonlinear magneto-optics even when both constituent materials have no cubic magneto-optical nonlinearity.

The space-averaged fields and displacements for a composite with metallic volume fraction f_1 are related by an equation of the same form as Eq. (6):

$$\langle \boldsymbol{D} \rangle = \varepsilon_{eff} \langle \mathbf{E} \rangle = e_{eff}^{(0)} \langle \mathbf{E} \rangle + \chi_{eff}^{(3)} |\langle \boldsymbol{E} \rangle|^2 \langle \boldsymbol{E} \rangle.$$

The diagonal and non-diagonal elements of the effective dielectric function ε_{eff} can be written as

$$\varepsilon_{eff}^{d} = F(\varepsilon_1^d,\ \varepsilon_2^d, f_1);\ \varepsilon_{eff}^{od} = \Phi\,(\varepsilon_1^d,\ e\varepsilon_2^d,\ \varepsilon_1^{od},\ \varepsilon_2^{od}, f_1), \quad (8)$$

where F and Φ are well known functions in the linear limit of EMA [25-27]. We expand these functions in a Taylor series about $\varepsilon_{eff}^{d(0)}$ and $\varepsilon_{eff}^{od(0)}$ taking into account that $\varepsilon_i = \varepsilon_i^{(0)} + \chi_i^{(3)} \langle |\boldsymbol{E}_i|^2 \rangle$, where $\langle |\boldsymbol{E}_i|^2 \rangle$ is the mean square of the electric field in the ith component in the linear limit [25-27]. Since within the linear limit [25-27]

$$f_i \langle |\boldsymbol{E}_i|^2 \rangle / \mathbf{E}_0^2 = (\partial\, \varepsilon_{eff}^d / \partial \varepsilon_i^d)_0 = F_1'\,(\varepsilon_1^{d(0)},\ \varepsilon_2^{d(0)}, f_1) = F_i', \quad (9)$$

where $\mathbf{E}_0$ is the external field, we have as a result:

$$\varepsilon_{eff}^{d} = \varepsilon_{eff}^{d(0)} + \chi_{eff}^{d} \boldsymbol{E}_0^2;\ \varepsilon_{eff}^{d} = \varepsilon_{eff}^{od(0)} + \chi_{eff}^{od}\, \boldsymbol{E}_0^2, \quad (10)$$

where

$$\chi_{eff}^{d} = f_1 K_1 \chi_1^d + f_2 K_2 \chi_2^d, \quad (11)$$

$$\chi_{eff}^{od} = f_1 Q_1 \chi_1^d + f_2 Q_2 \chi_2^d + f_1 M_1 \chi_1^{od} + f_2 M_2 \chi_2^{od}, \quad (12)$$

$$K_i = \frac{1}{f_i^2} F_i' |F_i'| = \frac{1}{f_i^2} \frac{\partial \varepsilon_{eff}^{d(0)}}{\partial \varepsilon_i^{d(0)}} \left(\frac{\partial \varepsilon_{eff}^{d(0)}}{\partial \varepsilon_i^{d(0)}} \right), \quad (13)$$

$$Q_i = \frac{1}{f_i^2} \frac{\partial \Phi}{\varepsilon_i^{d(0)}} |F_i'|, \quad (14)$$

$$M_i = \frac{1}{f_i^2} \frac{\partial \Phi}{\varepsilon_i^{od(0)}} |F_i'|. \quad (15)$$

The Eq. (11) describes cubic nonlinear optics and itis the same as the result of Refs. [25-27]. The Eq. (12) describes cubic nonlinear magneto-optics in nanocomposites. The coefficients K, Q and M are enhancement factors. They strongly depend on metallic volume fraction $f = f_1$ and frequency w. We calculated these coefficients using three different EMA schemes developed for linear magneto-optics [7], namely, Maxwell-Garnett approximation, Bruggeman approximation (EMA), and symmetrised Maxwell-Garnett approximation [7]. The details of these tedious but straightforward calculations will be published elsewhere. Some of the results can be summarized as follows.

The coefficients K, Q, M are large only at some charactereristic frequencies for every composition. The characteristic frequencies are the same for both optics (K) and magnetooptics (Q, M). They are connected with plasmon-type resonances in metallic grains or near metal-insulator interfaces, as well as with resonances of optical conductivity [28]. Therefore, as a rule $K(w)$, $Q(w)$, $M(w)$ spectra have at least two peaks for every composition.

The spectra $K(w)$, $Q(w)$, $M(w)$ depend on the type of EMA approach used for calculation. It means that nonlinear effects are more sensitive to granular alloy microstructure in comparison with linear phenomena.

Nonlinear magnetooptics in composites can be observed even when both components have no cubic magneto-optical nonlinearity ($\chi_1^{od} = \chi_2^{od} = 0$) but at least one of them exhibits nonlinear optics.

The enhancement factor for cubic magneto-optical nonlinearity can be as large as 10^3 and is larger than that for cubic optical nonlinearity.

Two other features of cubic magneto-optical nonlinearity should also be mentioned. First, within the EMA, the enhancement factor of magnetooptics at visible wavelengths cannot exceed 10^4. By analogy with nonlinear optics, it is quite reasonable to expect that fractal structure of composites close to the percolation threshold can provide both an additional enhancement and a shift of resonance frequency. Second, since magnetooptical effects, for example transversal Kerr effect, are linear with non-diagonal dielectric function but also depend on its diagonal component, the total enhancement factor of nonlinearity for every Kerr or Faraday magneto-optical effect is a rather complicated function of both linear and nonlinear optical and magnetooptical parameters, light frequency and incident angle.

5. Conclusion

The huge, in comparison with traditional magneto-optical effects, values of MRE in metal-insulator granular alloys with tunnel-type magnetoresistance were observed at infrared wavelengths. It makes MRE very promising for magneto-optical applications.

MRE can be very large not only in reflection mode considered above but also in transmission mode. Such investigations are in progress.

The experimental data on MRE clearly indicate the existence of spin dependent tunnelling at high frequencies, at least up to near infrared region of spectra, that is of primary importance for all high frequency applications of devices based on tunnelling, and for spintronics. One can expect that two new features will influence MRE at higher frequencies, namely, frequency dependence of tunnelling probability and phonon-assisted tunnelling. Unfortunately, it is difficult to study because of at visible wavelengths MRE is masking by strong interband optical transitions.

It should be also noticed that the MRE investigations are in the very beginning and a good many features of MRE have not been studied yet. For example, there is no data on incident angle, light polarization and temperature dependencies of MRE.

The developed within EMA theory predicts at least three orders of magnitude enhancement of cubic nonlinear magnetoptics in metal-insulator alloys at some definite frequencies corresponding to plasmon-like excitations and resonances of optical conductivity. Therefore, magneto-optical response should depend on light intensity and it makes possible by studying nonlinear magnetooptics to determine characteristic resonance frequencies, enhancement of electrical field at resonance frequencies, as well as to test EMA.

We hope that our brief review will stimulate experimental study of linear and nonlinear magneto-optical properties in magnetic granular alloys.

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