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High-frequency spin-dependent tunnelling in magnetic nanocomposites: Magnetorefractive effect and magnetoimpedance

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Abstract

Since the dielectric permittivity is linear with frequency-dependent conductivity, high-frequency properties for any kind of magnetic materials with the high magnetoresistance depend on magnetization. It manifests as magnetorefractive effect (MRE) in the infrared region of spectrum and as magnetoimpedance (MI) in the frequency range between radio and microwaves. The main mechanism of both MRE and MI in nanocomposites with tunnel-type magnetoresistance is high-frequency spin-dependent tunnelling. We report on recent results of theoretical and experimental investigations of MRE and MI in nanocomposites $Co_{51.5}Al_{19.5}O_{29}$, $Co_{50.2}Ti_{9.1}O_{40.7}$, $Co_{52.3}Si_{12.2}O_{35.5}$ and $(Co_{0,4}Fe_{0,6})_{48}(MgF)_{52}$. Most of the obtained experimental data for MRE and MI are consistent with the theory based on considering the tunnel junction between adjacent granules in percolation cluster as a capacitor.

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1. Introduction

Much interest in DC and AC spin-polarized transports in magnetic nanostructures has been triggered by the high DC magnetoresistance observed in all-metal and metal/oxides multilayers and granular alloys. High-frequency properties of magnetic materials are determined by the dielectric permittivity $\varepsilon(\omega)$ and magnetic permeability $\mu(\omega)$. Since the dielectric permittivity $\varepsilon(\omega)$ is linear with frequency-dependent conductivity $\sigma(\omega)$,

$$\varepsilon(\omega) = \varepsilon_r - i\,\frac{\sigma(\omega)}{\omega}, \quad (1)$$

where ε_r is the dielectric permittivity connected with the displacement current, high-frequency properties of any kind of magnetic materials with

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a high DC magnetoresistance depend on magnetization. It manifests as the magnetorefractive effect (MRE) in the infrared region of spectrum [1–3] and as the magnetoimpedance (MI) in the frequency range between radio and microwaves [4–6]. The MRE consists of changes in optical properties of systems with a high magnetoresistance when they are magnetized. This effect was studied thoroughly for GMR all-metal multilayers [1] and granular alloys [7] as well as for metal–insulator nanogranular alloys with tunnel-type magnetoresistance [2,3]. The MI (or high-frequency magnetoresistance) of all-metal multilayers was also studied in detail [4–6]. However, high-frequency properties of magnetic systems with tunnel junctions are far from being well understood [8,9]. For the best of our knowledge, the MI of metal–insulator nanocomposites has not been studied at high frequencies. Moreover, in spite of both the MRE and MI in systems with spin tunnelling junctions are due to high-frequency spin-dependent tunnelling, no comparative study of the MRE and MI for the same samples has been done.

In this paper, we present the result of such a comparative study of the MRE and MI in magnetic nanocomposites with tunnel-type magnetoresistance. The most of obtained experimental data for the MRE and MI are consistent with the theory based on considering the tunnel junction between adjacent granules in a percolation cluster as a capacitor.

2. Experiments

The objects of investigations were nanocomposite films with a metal volume fraction close to the percolation threshold. Nanogranular ferromagnetic metal–insulator thin films $Co_{51.5}Al_{19.5}O_{29}$, $Co_{50.2}Ti_{9.1}O_{40.7}$, $Co_{52.3}Si_{12.2}O_{35.5}$ and $(Co_{0,4}Fe_{0,6})_{48}(MgF)_{52}$ were fabricated by the method of tandem rf magnetron sputtering from various targets in argon and oxygen atmosphere onto uncooled glass substrates. The ferromagnetic granular size varied from 2 to 5 nm. A detailed description of the sample fabrication procedure as well as the methods and results of measurements of chemical, structural, electrical, magnetic and magneto-transport properties of the samples are given in Refs. [10,11]. The thin films thickness d, resistivity $\rho(H=0)$, magnetoresistance

$$\frac{\Delta\rho}{\rho} = \frac{\rho(H=0) - \rho(H)}{\rho(H=0)}, \tag{2}$$

are given in Table 1.

The optical reflection R and the MRE in reflection mode,

$$\frac{\Delta R(\omega, H)}{R} = \frac{R(\omega, H=0) - R(\omega, H)}{R(\omega, H=0)}, \tag{3}$$

were measured in the frequency band from 500 to 5000 cm^{-1} by using a FTIR PU9800 commercial Fourier spectrometer [2]. Optical reflection and MRE measurements were made for angles of incidence φ between 5° and 50° in a magnetic field of strength up to 1.5 kOe at room temperature.

High-frequency properties of the samples were studied in the extra high-frequency band (30–50 GHz) by placing the samples in the Open Resonator, described in detail in Ref. [12]. We measured the transmission coefficient $D(\omega, H)$ and its relative change

$$\frac{\Delta D}{D} = \frac{D(H=0) - D(H)}{D(H=0)} \tag{4}$$

Table 1
Compositions of the samples, magnitudes of the specific resistance ρ, the MR magnitude at $H = 1.5$ kOe, the magnetization-induced changes in the transmissivity D at $n = 44$ GHz, $H = 1.5$ kOe, and also MRE magnitude at $H = 1.5$ kOe for wave number, where MRE is maximum

Composition of samples, vol%	d, μm	ρ, μΩ cm	$\Delta\rho/\rho$ (%)	$\Delta D/D$(%) $\nu = 44$ GHz	$\zeta = \Delta R/R$(%) (ν, cm^{-1})
$Co_{51.5}Al_{19.5}O_{29}$	1.91	2.9×10^5	5.08	2.28	−0.9 (1100)
$Co_{50.2}Ti_{9.1}O_{40.7}$	2.02	6.1×10^6	2.42	1.6	−0.7 (1030)
$Co_{52.3}Si_{12.2}O_{35.5}$	1.67	4.5×10^8	2.99	—	+0.7 (1300)
$(Co_{0,4}Fe_{0,6})_{48}(MgF)_{52}$	1.0	$\sim10^9$	1.32	—	−1.3 (1000)

in a magnetic field of strength up to 1.5 kOe. Since D is determined by the impedance of the system, the parameter $\Delta D/D$ characterizes the MI. We did not observe any FMR features in the same frequency band for $H<10\,\text{kOe}$. It means that in the case considered $\mu(\omega,H)=\mu_0$, where μ_0 is the magnetic permeability of vacuum.

The obtained results are summarized in Table 1.

3. Results and discussion

The samples under investigation exhibit the high resistivity and tunnel-type magnetoresistance (Table 1) because their compositions are very close to the percolation threshold. It means that only a few contacts between adjacent granules in the percolation cluster are responsible for DC and AC transport. Each contact between granules is a tunnelling junction [2], which can be thought of as a capacitor [8]. Thus, if the same tunnelling junction is responsible for both DC and AC current, the frequency-dependent conductivity of nanocomposites can be written as

$$\sigma(\omega,H)=\frac{1+\mathrm{i}\varepsilon_{\mathrm{ins}}\omega\rho(H)}{\rho(H)}, \tag{5}$$

where $\varepsilon_{\mathrm{ins}}$ is the dielectric constant of the insulator between granules. Eq. (5) is valid if the tunnelling probability does not depend on frequency that is expected if $\omega\tau\ll 1$, where τ is the tunnelling time [13]. Besides, we neglect phonon-assisted and impurity-assisted tunnelling, as well as resonant double-barrier tunnelling [3]. Also, we did not take into account possible effects of electron–electron interaction on magnetocapacitance [9]. Thus, Eq. (5) is valid only close to the percolation threshold and can be viewed as only the initial step for qualitative interpretation of high-frequency properties of nanocomposites.

Using Eqs. (1)–(5) and Fresnel formula the MRE for arbitrary incident angles, light polarizations and thickness of the sample is straight forwardly calculated. The case of semi-infinite samples was considered in Refs. [2,14] and the obtained theoretical results are consistent with the experiment [2,15]. In the present paper, we consider the case of the thin film sample on the substrate. Firstly, we determined the optical constants (refraction and extinction indices) of both the sample and the substrate from optical measurements. Secondly, using Eqs. (1)–(5), Fresnel formula, the field dependence of magnetoresistance and the thickness of the sample, we calculated the MRE without any adjustable parameters. The obtained experimental and theoretical results for the MRE in nanocomposites $Co_{51.5}Al_{19.5}O_{29}$ are shown on Fig. 1. The agreement is fairly good in the whole spectrum range. It confirms the proposed mechanism of the MRE, the strong influence of interference of light on the MRE spectrum, as well as that made on model assumptions are not crucial for semi-quantitative description of the MRE. It should be noted that

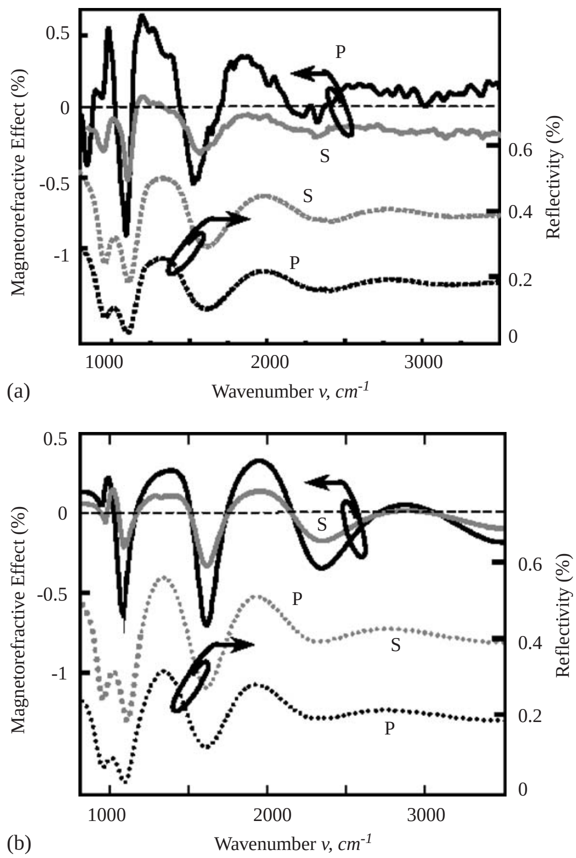

Fig. 1. Dispersion of MRE $\xi(\nu)$ (solid line) and $R(\nu)$ (dotted line) in nanocomposite $Co_{51.5}Al_{19.5}O_{29}$; $=1600\,\text{Oe}$; $\varphi=45^\circ$. (a) Experiment; (b) theoretical computation.

the MRE for all studied samples is at least two orders of magnitude larger than the traditional magneto-optical effects and can be observed only in the systems with a high magnetoresistance independent of their resistivity (Table 1).

Fig. 2 shows experimental data on the field dependence of both the parameters of the MI $\Delta D/D$ and magnetoresistance $\Delta\rho/\rho$ for nanocomposites $Co_{51.5}Al_{19.5}O_{29}$ and $Co_{50.2}Ti_{9.1}O_{40.7}$. The observed strict correlation between $\Delta D/D$ and $\Delta\rho/\rho$ is a direct evidence of the proposed mechanism of MI. In spite of all studied samples exhibit rather large MRE and magnetoresistance, we observed magnetization-induced changes in the transmissivity D at 30-50 GHz only for nanocomposites $Co_{51.5}Al_{19.5}O_{29}$ and $Co_{50.2}Ti_{9.1}O_{40.7}$ (Table 1). To explain this puzzling behaviour, namely, the high MI ratio for $Co_{51.5}Al_{19.5}O_{29}$, and $Co_{50.2}Ti_{9.1}O_{40.7}$, and insignificant MI for $Co_{52.3}Si_{12.2}O_{35.5}$, and $(Co_{0,4}Fe_{0,6})_{48}(MgF)_{52}$, let us consider the transmission of the three-layer system "air– nanocomposite–semi-infinite substrate" in the framework of the above-discussed model. For simplicity, we assume that the dielectric permittivity of the substrate and ε_r are equal to the dielectric permittivity of free space ε_0.

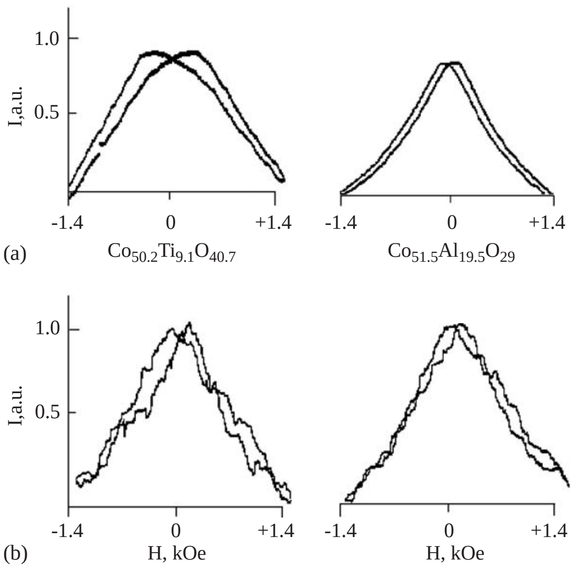

Fig. 2. The field dependence of the parameter of the MI $\Delta D/D$ (a) at $v = 44\,GHz$ and magnetoresistance $\Delta\rho/\rho$ (b) for nanocomposites $Co_{51.5}Al_{19.5}O_{29}$ and $Co_{50.2}Ti_{9.1}O_{40.7}$.

Then the transmission coefficient of this three-layer system is [6]

$$D = \frac{2Z_2Z}{2Z_2Z ch\, k_2d + (Z_2^2 + Z^2) sh\, k_2d}, \tag{6}$$

where $Z = (\mu_0/\varepsilon_0)^{1/2}$ is the impedance of free space, $Z_2 = (\mu_0/\varepsilon_0)^{1/2}$ is the impedance of the nanocomposite thin film, $k_2 = i\omega(\varepsilon\mu_0)^{1/2}$. For higly conductive metals the second term in Eq. (1) is many orders of magnitide larger that the first term. But it is not valid for nanocomposites close to the percolation threshold (Table 1). In contrast, for these systems the second term in Eq. (1) is of the same order of magnitude as the first one and even larger. For simplicity we consider the limit case then $\sigma(\omega)/\omega\varepsilon \gg 1$. Then it immediately follows from Eqs. (1)–(6) that

$$\frac{\Delta D}{D} = \frac{D(H=0) - D(H)}{D(H=0)} = \frac{1}{2}\frac{d}{c}\frac{1}{\varepsilon_r\rho}\frac{\Delta\rho}{\rho}. \tag{7}$$

Eq. (7) allows us to explain qualitatively the main features of the MI in nanocomposites. Similar to the MRE $\Delta D/D$ is linear with $\Delta\rho/\rho$ but, in addition, the MI is inversely proportional to ρ. Since the resistivity ρ for $Co_{52.3}Si_{12.2}O_{35.5}$ and $(Co_{0,4}Fe_{0,6})_{48}(MgF)_{52}$ is several orders of magnitude larger than that for $Co_{51.5}Al_{19.5}O_{29}$, and $Co_{50.2}Ti_{9.1}O_{40.7}$ (Table 1), the MI for first two systems is negligible in comparison with that for last two systems. The MI parameter for $Co_{50.2}Ti_{9.1}O_{40.7}$ is less than that for $Co_{51.5}Al_{19.5}O_{29}$ because the resistivity is larger and the magnetoresistance is smaller for $Co_{50.2}Ti_{9.1}O_{40.7}$ (Table 1). At last, for parameters listed in Table 1 Eq. (7) gives the MI ratio of 0.66% for $Co_{51.5}Al_{19.5}O_{29}$ that is approximately three times smaller than the experimental value. Taking into account assumptions made above ($\sigma(\omega)/\omega\varepsilon \gg 1$, $\varepsilon_r = \varepsilon_0$) the agreement is wholly satisfactory. Besides, we neglected a contribution to the MI connected with $\mu(\omega, H)$ putting in Eqs. (6),(7) $\mu(\omega, H) = \mu_0$. For quantitative description one should also consider the finite dimension and thickness of the substrate as well as the difference between the dielectric permittivity of air, nanocomposite and substrate.

4. Conclusion

The experimental data on the MRE and MI in metal–insulator nanocomposites clearly indicate the existence of spin-dependent tunnelling at high frequencies, at least up to near-infrared region of spectra. Both effects are due to high-frequency magnetoresistance and can be explained in the framework of the simple model based on the representation of the tunnel junction between adjacent granules in the percolation cluster as a capacitor. In the framework of this model, the MI in transmission mode cannot be observed in insulating nanocomposites that are in agreement with the experiment.

The magnitude of both the MRE and the MI in nanocomposites with a high magnetoresistance is about 1%. The huge, in comparison with traditional magnetooptical effects, values of the MRE makes the MRE very promising for magnetooptical applications. The high values of the MI in nanocomposites at 30–50 GHz, observed for the first time, clearly indicate that the MI can be observed at very high frequencies. It makes it possible to design magnetic field controlled devices, operating in a wide frequency band, including microwaves.

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