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Microwave-Frequency Spin-Dependent Tunneling in Nanocomposites

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Abstract—The transmission coefficient of films of $Co_{51.5}Al_{19.5}O_{29}$, $Co_{50.2}Ti_{9.1}O_{40.7}$, $Co_{52.3}Si_{12.2}O_{35.5}$, and $(Co_{0.4}Fe_{0.6})_{48}(MgF)_{52}$ ferromagnetic metal–insulator magnetic nanocomposites exhibiting tunneling magnetoresistance and the magnetorefractive effect for electromagnetic waves was studied in the frequency range 30–50 GHz. The transmission coefficient of the first two compositions varies strongly under an applied magnetic field, and its variation exhibits a linear correlation with the field dependence of magnetoresistance. For the other two compositions, the transmission coefficient does not depend on magnetic field. The data obtained are interpreted in terms of the concept of microwave spin-dependent tunneling. © *2005 Pleiades Publishing, Inc.*

The high-frequency properties of metallic multilayers with giant magnetoresistance have been studied over a fairly broad frequency range (see [1–6] and references therein). Magnetization of multilayers gives rise not only to a substantial reduction in resistivity but also to a change in the permittivity. As a consequence, the impedance and optical properties of multilayers become dependent on magnetic field. In the optical range, this phenomenon was termed the magnetorefractive effect [6], and in the radio-frequency and microwave ranges, high-frequency magnetoresistance or magnetoimpedance [1–5]. Similar effects are also expected to occur in systems with a considerable magnetoresistance of any type, including systems with tunneling magnetoresistance (TMR). Ferromagnetic metal–insulator nanocomposites with a metal content near the value corresponding to the percolation threshold, as well as magnetic trilayer and multilayer systems with tunneling barriers, belong to TMR systems. Investigation of the microwave properties of these systems will make it possible to determine the mechanisms of spin-dependent tunneling, establish the frequency dispersion of conductivity, check the recent concept of magnetocapacitance [7], determine the possible operational range of spintronic devices based on TMR systems (for example, spin filters, magnetic storage devices, magnetic sensors), and suggest new possible areas of their application [6]. Recent studies of the magnetorefractive effect (MRE) in reflection performed in nanocomposites in the near IR region [6, 8, 9] have confirmed the existence of spin-dependent tunneling up to optical frequencies. The present paper reports on an experimental study of the transmission of millimeter-range electromagnetic waves (30–50 GHz) through films of ferromagnetic metal–insulator magnetic nanocomposites possessing TMR.

As subjects for the study, we chose films of nanocomposites of various compositions with metal contents in the immediate vicinity of the percolation threshold. The techniques employed in the preparation and structural characterization of the samples are described in [8, 9]. The composition, film thickness d, residual electrical resistivity ρ ($H = 0$), TMR parameter $\Delta\rho/\rho = \frac{\rho(H=0)-\rho(H)}{\rho(H=0)}$, and MRE parameter in reflection $\zeta = \Delta R/R = \frac{R(H=0)-R(H)}{R(H=0)}$ measured in a field $H = \pm 1.5$ kOe, as well as the relative changes in the transmission coefficient $\frac{\Delta D}{D} = \frac{D(H=0)-D(H)}{D(H=0)}$ in the same field $H = \pm 1.5$ kOe at 44 GHz, are listed in the table. Because the reflectance R at optical frequencies and the MRE depend strongly on the light frequency ν, the table specifies, for each composition, the maximum values of the MRE at the corresponding frequency, which were taken from [8–10]. Also presented are the values of TMR in a field of ±10 kOe; for simplicity, we neglect a certain difference between the values of the corresponding parameters in a zero external magnetic field and in the state with zero magnetization (which occurs in a field equal to the coercive force). All measurements were conducted at room temperature with an in-plane field.

The microwave transmission coefficient D of nanocomposites in the range 30–50 GHz and its relative

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Parameters of the films under study

No.	Sample composition, vol %	d, μm	Δρ/ρ, %		$\Delta D/D$, %	ρ, μΩ cm	$\zeta = \Delta R/R$, % (ν, cm^{-1})
			$H = \pm 10$ kOe	$H = \pm 1.5$ kOe			
1	$Co_{51.5}Al_{19.5}O_{29}$	1.91	9.2	5.08	2.28	2.9×10^5	−0.9 (1100)
2	$Co_{50.2}Ti_{9.1}O_{40.7}$	2.02	5.8	2.42	1.6	6.1×10^6	−0.7 (1030)
3	$Co_{52.3}Si_{12.2}O_{35.5}$	1.67	4.1	2.99	Not detected	4.5×10^8	+0.7 (1300)
4	$(Co_{0.4}Fe_{0.6})_{48}(MgF)_{52}$	1	13	1.32	″	$\sim 10^9$	−1.3 (1000)

change under magnetization, $\Delta D/D$, which can be called the magnetoimpedance parameter in transmission, were measured using the open-resonator technique described in detail in [5]. It should be stressed that, in contrast to the optical reflectance or transmittance, the transmission coefficient relates to the ratio of the wave amplitudes rather than the ratio of the intensities. We also studied ferromagnetic resonance (FMR) in our samples in fields of up to 20 kOe. We note that, in the frequency range 30–50 GHz, the FMR is observed to exist only above 8 kOe, i.e., in fields considerably stronger than those in the case of $\Delta D/D$ measurements.

The measured values of $\Delta D/D$ suggest several conclusions. First, the $Co_{51.5}Al_{19.5}O_{29}$ and $Co_{50.2}Ti_{9.1}O_{40.7}$ nanocomposites exhibit considerable changes in the transmission coefficient under magnetization, with the magnetoimpedance parameter $\Delta D/D$ being of the order of the TMR. A comparison of the field dependences of $\Delta D/D$ and TMR (see Fig. 1) argues convincingly for the observed effect being (as should be expected) a frequency analog of the TMR. Because no FMR is observed in our samples in the frequency range 30–50 GHz in fields of up to 1.5 kOe, one can also conclude that this effect is not related to the dependence of magnetic per-

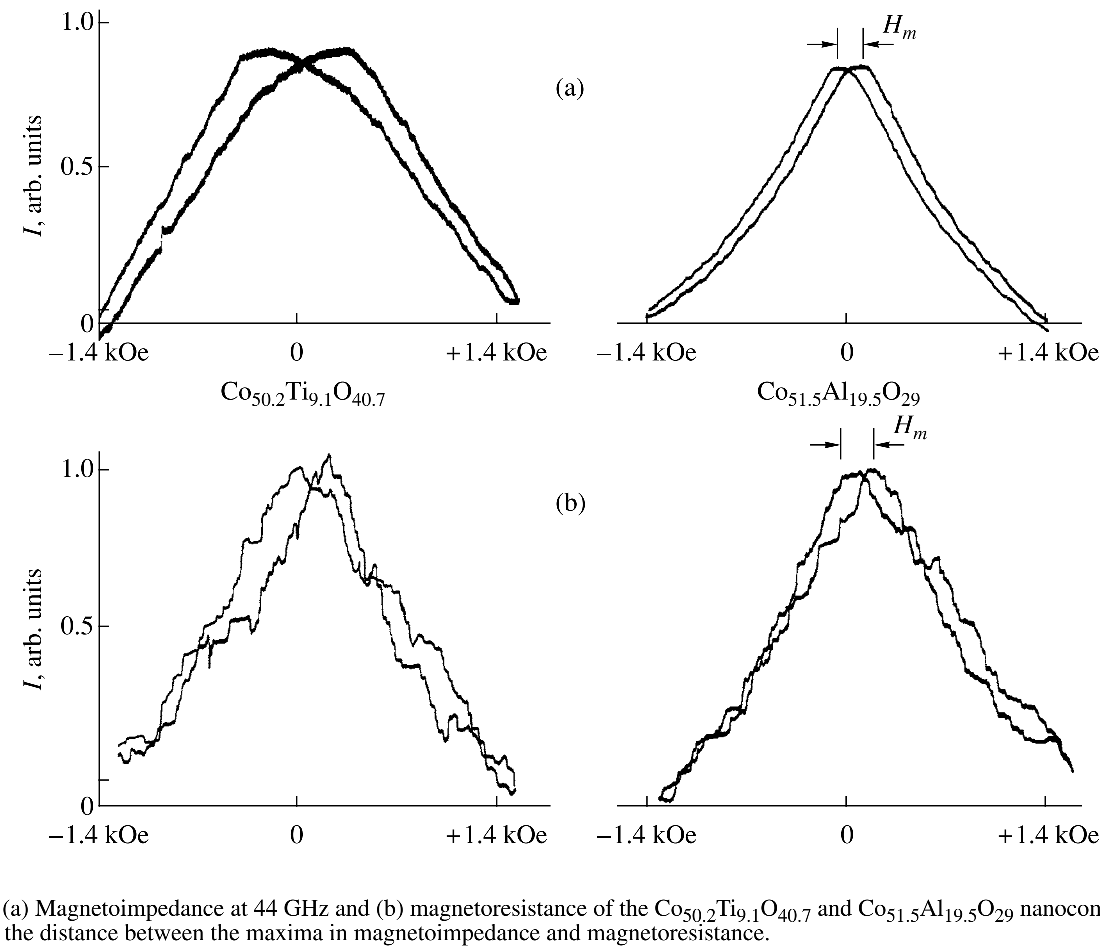

Fig. 1. (a) Magnetoimpedance at 44 GHz and (b) magnetoresistance of the $Co_{50.2}Ti_{9.1}O_{40.7}$ and $Co_{51.5}Al_{19.5}O_{29}$ nanocomposites. $2H_m$ is the distance between the maxima in magnetoimpedance and magnetoresistance.

meability on magnetic field. Second, the magnetoimpedance $\Delta D/D$ for these two compositions is only weakly frequency-dependent (unlike the MRE). This feature correlates with the magnetoimpedance measurements carried out on metallic multilayers [4] and corroborates the interference nature of the strong dependence of the MRE on the frequency of light [8]. Third, the magnetoimpedance parameter for these samples turns out to be larger than the MRE. Finally, no magnetoimpedance was detected in samples with the compositions $Co_{52.3}Si_{12.2}O_{35.5}$ and $(Co_{0.4}Fe_{0.6})_{48}(MgF)_{52}$, which exhibit appreciable MRE and TMR; this result, at first glance, is at odds with the concept of the magnetoimpedance being a frequency analog of the TMR.

Let us show that these features of magnetoimpedance do find explanation within a simple model. In calculating the impedance, we have to take into account that, near the percolation threshold ($x \approx x_c$), the electrical resistivity of nanocomposites exceeds the resistivity of conventional metals by 7 to 10 orders of magnitude and increases strongly (by a few orders of magnitude) when crossing over from metallic conduction ($x \geq x_c$) to tunneling and hopping transport ($x \leq x_c$).

As in [4], we neglect the difference between the wave impedance of the insulating substrate and that of free space $Z = (\mu_0/\varepsilon_0)^{1/2}$. Under these conditions, the transmission coefficient of the air–film–air trilayer system can be written as

$$D = \frac{2Z_2Z}{2Z_2Z\cosh k_2d + (Z_2^2 + Z^2)\sinh k_2d}, \quad (1)$$

where $Z_2 = (\tilde{\mu}/\tilde{\varepsilon})^{1/2}$ is the impedance of the nanocomposite and $k_2 = i\omega(\tilde{\varepsilon}_2\tilde{\mu}_2)^{1/2}$ is the wavenumber. For microwave frequencies and far from the FMR region, we can assume the magnetic permeability of the nanocomposite to be $\tilde{\mu}_2 = \mu_0$ and, in the expression for the complex permittivity

$$\tilde{\varepsilon}_2 = \varepsilon_2 - i\frac{\sigma(\omega)}{\omega} \quad (2)$$

the second term is of the order of or less than the first for the high-resistivity systems under study here (see table). Considering the limiting case where $\sigma(\omega)/\omega\varepsilon_2$ is a small parameter, we obtain from Eq. (1)

$$D = \exp\left[-i\frac{\omega}{c}d - \frac{\omega}{c}d\frac{\sigma(\omega)}{2\omega\varepsilon_2}\right] \approx \exp\left[-i\frac{\omega}{c}d\right]\left(1 - \frac{\omega}{c}d\frac{\sigma(\omega)}{2\omega\varepsilon_2}\right), \quad (3)$$

$$\frac{\Delta D}{D} = \frac{D(H) - D(H=0)}{D(H=0)} = \frac{1}{2}\frac{d}{c}\frac{1}{\varepsilon_2\rho}\frac{\Delta\rho}{\rho}, \quad (4)$$

where we neglected the possible dependence of the conductivity on frequency and set $\sigma(\omega, H) = 1/\rho(H)$. It should be pointed out that the above approximations are too rough to be used in a quantitative description, because the parameter $\sigma(\omega)/\omega\varepsilon_2$ is not small for the first two compositions in the table. We also cannot disregard the frequency dependence of the conductivity (see discussion of the possible frequency dependence of the conductivity of magnetic composites in [6]). Nevertheless, Eq. (4) offers an interpretation for the observed relations. As follows from Eq. (4), the magnetoimpedance parameter $\Delta D/D$ and the TMR are linearly correlated. Furthermore, the magnetoimpedance is inversely proportional to the sample resistivity ρ. For the last two samples in the table, which are located on the insulating side of the percolation transition, the resistivity exceeds that of the first two samples by about four orders of magnitude. Therefore, the parameter $\Delta D/D$ for the former samples is negligible, despite the fact that the values of MRE and TMR for them are higher. Equation (4) also shows that the parameter $\Delta D/D$ is of the same sign as the TMR and is not larger than the TMR. All these conclusions fit the results displayed in the figure and in the table. The frequency dependence of the conductivity and of the magnetoimpedance parameter of magnetic nanocomposites over a broader frequency range, as well as the problem of quantitative description of the experiment, will be treated in a future publication.

We can conclude that the giant magnetoimpedance of nanocomposites at 30–50 GHz is a consequence of microwave spin-dependent tunneling and is observed only in compositions on the metallic side of the percolation transition.

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REFERENCES

1. J. J. Krebs, P. Lubitz, A. Chaiken, and G. A. Prinz, J. Appl. Phys. **69** (8), 4795 (1991).
2. A. B. Drovosekov, N. M. Kreĭnes, D. I. Kholin, V. F. Meshcheryakov, M. A. Milyaev, L. N. Romashev, and V. V. Ustinov, Pis'ma Zh. Éksp. Teor. Fiz. **67**, 690 (1998) [JETP Lett. **67**, 727 (1998)].
3. A. B. Rinkevich, L. N. Romashev, and V. V. Ustinov, Zh. Éksp. Teor. Fiz. **117** (5), 960 (2000) [JETP **90**, 834 (2000)].
4. V. V. Ustinov, A. B. Rinkevich, L. N. Romashev, A. M. Burkhanov, and E. A. Kuznetsov, Fiz. Met. Metalloved. **96** (3), 52 (2003) [Phys. Met. Metallogr. **96**, 291

(2003)]; Fiz. Tverd. Tela (St. Petersburg) **37** (2), 561 (1995) [Phys. Solid State **37**, 306 (1995)].

5. D. P. Belozorov, V. N. Derkach, S. N. Nedukh, A. G. Ravlik, S. T. Roschenko, I. G. Shipkova, S. I. Tarapov, F. Yidiz, and B. Aktas, J. Magn. Magn. Mater. **263** (3), 315 (2003).
6. A. Granovsky and M. Inoue, J. Magn. Magn. Mater. **272–276** (Suppl. 1), E1601 (2004).
7. S. T. Chui and Liangbin Hu, Appl. Phys. Lett. **80** (2), 273 (2002).
8. A. B. Granovsky, I. V. Bykov, E. A. Gan'shina, V. S. Gushchin, M. Inoue, Yu. E. Kalinin, A. A. Kozlov, and A. N. Yurasov, Zh. Éksp. Teor. Fiz. **123** (6), 1256 (2003) [JETP **96**, 1104 (2003)].
9. A. Granovsky, V. Gushchin, I. Bykov, A. Kozlov, N. Kobayashi, S. Ohnuma, T. Masumoto, and M. Inoue, Fiz. Tverd. Tela (St. Petersburg) **45** (5), 868 (2003) [Phys. Solid State **45**, 911 (2003)].
10. I. V. Bykov, E. A. Gan'shina, A. B. Granovsky, V. S. Gushchin, A. A. Kozlov, T. Masumoto, and S. Ohnuma, Fiz. Tverd. Tela (St. Petersburg) **47** (2), 268 (2005) [Phys. Solid State **47**, 281 (2005)].

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