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Extra high frequency features of nanogranular magnets with GMI

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Abstract

The results of research of electromagnetic waves interaction with the promising magnetic nanogranular layered structures in (10, 30–45) GHz frequency bands are presented. We applied the electron spin resonance (ESR) technique to study the magnetic order of nanostructures, which demonstrate the giant magnetic resistance (GMR) effect and the extra high frequency giant magnetic impedance (GMI) effect. The data under discussion are the ESR/FMR-linewidth, -line positions and their variations depending on the temperature. Besides this, the ESR response from the specimen, located in plane or normally to the external magnetic field, has been detected and the magnetization of saturation has been calculated using these data. The nanogranular Co- and Fe-contained specimens, which exhibit the GMI effect (at 30–45 GHz), have been studied. The correlation between the magnetic structure of specimens and GMI magnitude is considered.

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1. Introduction

Nanogranular magnets are of great interest today due to the fact that these artificial materials are very promising structures for applications in spin-electronics [1] of extrahigh frequency band [2] and infrared band [3,5].

The novelties of the presented results are the following: Firstly, the specimens exhibit a pronounced magnitude of giant magnetic resistance/impedance (GMR/GMI) phenomena that makes them prospective for application as electronically controlled circuit components in extra high frequency band. Namely, the magnitudes of GMI approx. 2–4% have been detected [4,5] for frequencies 30–45 GHz. Secondly, one of the most important tasks in this direction is the definition of the main types of interactions between the magnetic granules and atoms, which are responsible for GMI effect in the extra high frequency band. This should help to increase the magnitude of GMI effect. It is very important from the point of view of its technological application. Thirdly, the method of electron spin resonance (ESR), applied here for the extra high frequency studies, is very informative for study of electron spin system features, which are responsible for the formation of magnetic order in the nanocomposite.

In the given paper, the analysis of results of ESR experiments in the wide temperature range and at extra high frequencies close to frequencies of the prospective application is given in order to find the correlation between the magnetic order in the nanostructure and the GMI effect magnitude.

2. Experimental

The special experimental cell designed as an open quasioptical resonator [4] has been used in extra high frequency ESR technique. Such kind of resonator provides positioning of the specimen in the area of the maximum of either electrical or—magnetic components of the electromagnetic field without dismantling of the resonator cell.

We carried out a comparative analysis of ESR data from Co- and Fe-containing specimens (performed in various

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laboratories) namely: $(Co_x(Al_2O_3)_{1-x} (x = 21.0\%; 26.0\%; 32.8\%; 38.8\%; 43.4\%); Co_x(TiO)_{1-x} (x = 31.6\%; 41.4\%; 49.4\%; 57.6\%); (Co-Fe-Zr)_x(SiO_2)_{1-x} (x = 34\%; 40\%; 43\%; 47\%)$ and some other samples of the same composition manufactured by the similar technique. The specimens were shaped as thin films with the thickness $1-3 \mu m$ and with the size of each granule of order 3-5 nm. Let us note that the GMR/GMI effect, which is caused in these specimens by the tunneling of polarized electrons, is called the tunnel magnetic resistance/impedance (TMR/TMI) [3] as well.

The experimental resonance frequency-resonance field dependencies were approximated by the known Kittel formula in order to find the magnetization of saturation value. This formula for the "parallel" geometry of experiment (magnetic component of extra-high frequency field is orthogonal to the static magnetic field vector and both of them lie in the specimen plane) is

$$v_{\rm res}^{\rm II} = \frac{g\beta}{h} \sqrt{H_{\rm res}^{\rm II}(H_{\rm res}^{\rm II} + 4\pi M_{\rm S})} \tag{1}$$

and for the "perpendicular" geometry (the magnetic component of extra-high frequency field lies in the specimen plane and the static magnetic field vector is directed normally to the sample plane) is

$$v_{\rm res}^{\perp} = \frac{g\beta}{h} \left(H_{\rm res}^{\perp} - 4\pi M_{\rm S} + H_{\rm an}^{\rm eff} \right),\tag{2}$$

where $v_{\rm res}$ is the resonant frequency; g is the spectroscopic splitting factor (equals to $2.05 \pm 5\%$; usually [4]), β is the Bohr magneton; h is the Plank constant; $H_{\rm res}$ is the resonant magnetic field; $M_{\rm S}$ is the magnetization of saturation. The $H_{\rm an}^{\rm eff}$ is an effective field, which describes the energy of disordering of electron spin system.

The magnetization saturation, which is calculated in such way, is presented in Fig. 1. One can see easily that the

800 600 Magnetization, G 400 ⊠ ⊠ Co_v(Al₂O₃)_{1-x} 200 ⊙ (Co-Fe-Zr)_v(SiO₂)₁ \triangle Co_v(TiO₂)_{1-x} 0 20 30 40 50 60 X, %

Fig. 1. Magnetization of saturation obtained from the ESR data versus the concentration of Co.

magnetization grows with the concentration of Co. This fact seems to be natural. But the extrapolation of the data obtained for the concentration x = 100% gives the value of magnetization much less than the value for the bulk material (Co or Fe). This fact, which is observed for all samples, is caused undoubtedly by the influence of the shape of separate granules on the interaction between granules.

Just as well, the linewidth dependence on the concentration for the specimens mentioned (Fig. 2) has the monotonous character. It is well known that the width of the ESR line accompanies the magnetic ordering (the collinearity) of the electron spin system. Thus it is easy to make a conclusion that the increasing of Co-concentration leads to establishing of more collinear electron spin system in the nanostructure.

On the other hand let us note that the maximum of GMI effect is observed in the vicinity of the concentration $x \approx 44\%$ ($\Delta T/T \approx 2-3\%$ [5]), namely just for that concentration where the percolation from the dielectric phase to metallic one takes place. However, as one can see there is no any noticeable correlation between the linewidth and the magnetization on the one hand and the GMR/GMI magnitude on the other hand. Just as the magnetization, the linewidth varies monotonically in the vicinity of the percolation threshold, where the conductivity exhibits well-known jump-like behavior.

This fact indicates that not only the direct exchange interaction (and not the RKKY-exchange, which usually defines the magnetic order in the metal phase) is responsible for the magnetic order establishing. Most likely, it is the dipole–dipole interaction, which plays the main role in magnetic order forming, for both metal and dielectric phases.

In order to define the rigidity of magnetic order in nanogranular magnets we studied Co- and Fe-contained specimens with Co-concentrations close to the percolation



Fig. 2. Linewidth versus the Co-concentration for frequency 44 GHz.



Fig. 3. Linewidth for Co-contained granular structures depending on the temperature at 10 GHz for (a) "perpendicular" and (b) "parallel" orientation.

threshold. Namely, the temperature dependence of linewidth in 10 GHz band has been detected (Fig. 3).

It is easy to see that the linewidth grows drastically when the temperature falls up to $T \approx 5 \text{ K}$ for both parallel and perpendicular orientations of experiment. Moreover, for the lowest temperatures T = 4.2 K the linewidth increases to magnitudes which are comparable with resonance field magnitudes. This fact is the known sign of electron spin (magnetic) system becoming quite disordered (asperomagnetic, sperimagnetic, superparamagnetic, etc.).

It is easy to see also that the GMI magnitude is larger in those composites for which the linewidth variations with temperature damping are the smallest. Namely the largest GMI magnitudes were detected for Co–Al–O ($\Delta T/T \approx 2.28\%$) and for Co–Ti–O ($\Delta T/T \approx 1.6\%$).

This means that the magnetic order for these two nanostructures appears stronger than for others, i.e. the inter-spin connections are more resistant to the temperature action/fluctuation.

Besides let us note that the electron spin system exhibits non-collinear (most likely-superparamagnetic) magnetic order when the temperature decreases. The noncollinearity becomes more explicit at the lowest temperatures. Thus the effective anisotropy field H_{an}^{eff} can be estimated using Eqs. (1) and (2). This field can be considered as a result of the disordering energy in electron spin system and reaches values of order $H_{an}^{eff} \approx 5800 - 6500$ Oe at $T \approx 4.2-5$ K. Let note as well that the magnitude of H_{an}^{eff} coincides with data obtained for 75 GHz band [2].

3. Conclusions

The ESR-study of the set of GMR/GMI (TMR/TMI) magnetic nanostructures in the wide frequency band (10–45 GHz) has been performed for the first time. It gives the following outcomes:

- It has been revealed, that the more GMR/GMI effect magnitude in the electron spin system in the specimens under study is, the more steady/stable they are to the external impacts.
- The magnitude of the effective anisotropy field, which causes the electron spin system disorder, has been estimated.
- It is showed that the most probable origin of magnetic order in nanostructures is connected not with the direct exchange but with dipole–dipole interaction.

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