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ELECTRON SPIN RESONANCE PROPERTIES OF MAGNETIC GRANULAR GMI—NANOSTRUCTURES IN MILLIMETER WAVEBAND

S. Tarapov,[1] T. Bagmut,[1] A. Granovsky,[2] V. Derkach,[1] S. Nedukh,[1] A. Plevako,[1] S. Roschenko,[3] and I. Shipkova[3]

[1]*Institute of Radiophysics and Electronics NAS of Ukraine*
12 Ac. Proskura St.
Kharkov, 61085, Ukraine
E-mail: tarapov@ire.kharkov.ua
[2]*Faculty of Physics*
Moscow State University
119992, Moscow, Russia
[3]*National Technical University*
"Kharkov Polytechnical Institute"
21 Frunze St.
61002, Kharkov, Ukraine

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Abstract

The millimeter waveband giant magnetoimpedance (GMI) and magnetoresonance (FMR) properties of thin-layered magnetic nanostructures are analyzed experimentally for wide temperature range. The correlation between FMR and GMI features are under discussion. The manifestation of the anisotropy established by the millimeter waveband FMR experiments and possible reasons of anisotropy appearance are given.

Key words: millimeter waveband, electron spin resonance/ferromagnetic resonance (ESR/FMR) spectroscopy, Giant Magnetic Impedance/Resistance (GMI/GMR).

1. Introduction

Magnetic nanostructures that demonstrate the Giant Magnetic Impedance (GMI) phenomenon are promising objects for application in millimeter waveband technologies. This phenomenon as an AC-analog of Giant Magnetic Resistance (GMR) phenomenon [1] is one of the most interesting effects, which is very close to its technological implementation today. The situation is caused by the nature of the phenomenon, which shows up as a drastic increasing of the impedance for special magnetic nanostructures, while rather small static magnetic field is applied. According to the results of contemporary researches, the origin of GMI-phenomenon for EHF and RF bands is close to the GMR [2, 3] for the majority of nanostructure types. As it was determined in various publications, spin–dependent GMR/GMI processes are caused by strong exchange interaction between magnetic elements of nanostructures. These elements have the size of nanometer order and they are: nanolayers for multilayered structures [2-4] or nanoclusters for granular structures [5].
Structures that demonstrate the GMR phenomenon have already found their application today as the basis for the heads of hard-disks of super-high capacity, position sensors etc.. GMI phenomenon is studied much less due to primary the complexity of correct measuring technique for the EHF band.
But from the point of view of GMI applications, particularly for usage in millimeter waveband, the GMI structures can be used for design of such devices as electronically controlled polarizers, attenuators, scanning antennas etc., with very small switching/relaxation times ($\sim 10^{-12}$s [4]). Moreover, they should probably be of even more interest for higher frequencies [5], particularly for infrared band as the basis for devices similar to those above mentioned. Such kind of devices promises to be rather cheap and simple. The only problem, which should be solved on the way is creating the structures, that possess large enough GMI effect (20-50 %) at quite small magnetic field values (20-100 Oe).
It is necessary to note, that multilayered structures, which are well-ordered magnetic systems demonstrate today larger value of GMR (and GMI effect) effect than granular structures. But the advantage of the granular structures before multilayered ones is simpler and cheaper technology of their manufacturing.
The main aim of the given paper is to research granular magnetic nanostructures in millimeter waveband by the ferromagnetic resonance (FMR) technique. The research of nanostructures in the frequency band of their prospective application is of great interest now.

2. Specimens and Experimental Technique

The measurements of millimeter wave interactions with the granular magnetic metal-dielectric films:
#1 - $Co_{51.5}Al_{19.5}O_{29}$, #3 - $Co_{52.3}Si_{12.2}O_{35.5}$,
#2 - $Co_{50.2}Ti_{9.1}O_{40.7}$, #4 - FeCoMnF
possessing tunnel magnetoresistance have been carried out. All the samples have the concentrations of Co granules of such magnitude that the specimen's conductivity is in the vicinity of the threshold of percolation from metal to dielectric phase. For such

sample orientation/"geometry" the maximum amount of tunnel contacts between Co granules is achieved. On this reason one should expect a maximum value of GMR and GMI magnitude exactly for the given concentrations.
Granular ferromagnetic metal–insulator films with high magnetoresistance were prepared by the method of double RF-magnetron sputtering from various targets of Co-alloys in argon and oxygen atmosphere onto non-cooled glass substrates. The film thickness was 2μm and the size of Co grains varied from 2 to 5 nm. A detailed description of the sample preparation procedure as well as the methods and results of measurements of chemical, structural, electrical, and magnetic parameters of the samples are given in [6, 7].
We used two-mirror Fabri–Perot resonator for measurements of GMI and FMR as an adequate technique ([8] etc.) for 30-80 GHz band studies with digital data acquisition system. The FMR at T=300 K have been measured on the special ESR spectrometer of 30-50 GHz band. The resonator's modification used for T=300K measurements is shown in Fig.1a,b. The plane specimen has been placed on the lower plane mirror and fixed with EHF-field concentrator to protect the shifting of ferromagnetic specimens at large magnetic fields. Thus the EHF vector (h) and the static magnetic field (H) lie in the plane of the loser mirror. The loaded quality factor reaches values $3\text{-}6{\cdot}10^3$. For the measurements at low temperatures T=4.2 K the radiospectroscopy complex BURAN [8] has been used.

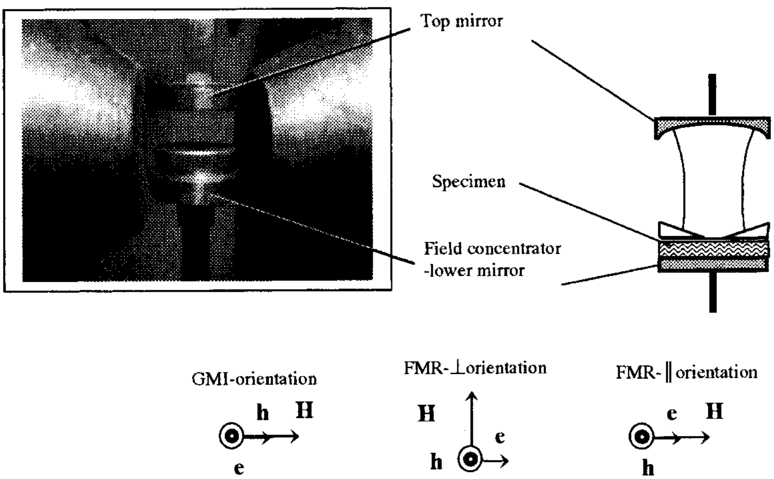

Fig.1. *Fabri-Perot resonator cell for granular nanolayered specimens research*

The magnetoimpedance (AC-resistance on frequencies 35-75 GHz) and magnetoresistance (DC-resistance) magnitudes have been measured as a function of the external static magnetic field (H). As well the ferromagnetic resonance (FMR) response has been detected and analyzed along with the same data for temperatures of T=300 K and T=4.2 K.

Magnetoresistance was measured by the conventional 4-point scheme [2, 3], where the external magnetic field was applied along the plane of the specimen and the DC (as well as AC) current was applied in the cross direction. The magnetoresistance is represented as "the relative magentoresistance ratio" in the following way

$$(\Delta R/R)_{max} = (R_{max}-R_s)/R_s \quad (1)$$

where R_{max} is the maximum of R in the range of the field applied, R_s is the magnetoresistance at $H \rightarrow \infty$. Magnetoimpedance magnitude for millimeter waveband was estimated via the transmission coefficient T of the Fabri-Perot resonator. The theory of perturbations approach could be applied here for small perturbations of the resonator with the specimen (as in our case). Thus one can suggest that the impedance magnitude Z should be proportional to T coefficient. At very small disturbance of electrodynamical system, when the disturbance theory is applicable the relative impedance can be estimated by the formula similar to (1):

$$(\Delta Z/Z)_{max} \approx (\Delta T/T)_{max} = (T_{max}-T_s)/T_s \quad (2)$$

The GMR and GMI data are presented in Table 1 col.2, 3 correspondingly. Let us note, that the GMI response is noticeable only for samples ##1, 2.
The registration of FMR signal was realized with the aid of automated data acquisition system designed by the authors. The kernel of the system is Atmel's AVR microcontroller AT90S2313 and two 12-bit Analog Digital Converters (ADC), which perform the data acquisition, conversation them to the digital signal and transmission further to personal computer (PC). Developed software allows one to accept experimental data, process them and to display on the screen of PC in real time mode. Such hardware/software complex allows us to detect the FMR signal with accuracy of magnetic field adjusting not less than 1 Oe in the static magnetic band, which corresponds to the typical linewidth $\Delta H_{1/2} \approx 4$ kOe.

3. Ferromagnetic Resonance Measurements

In order to establish the correlation between GMI response and the magnetization of the structure the FMR study were performed. The typical FMR detected with ADC unit is presented in Fig.2.

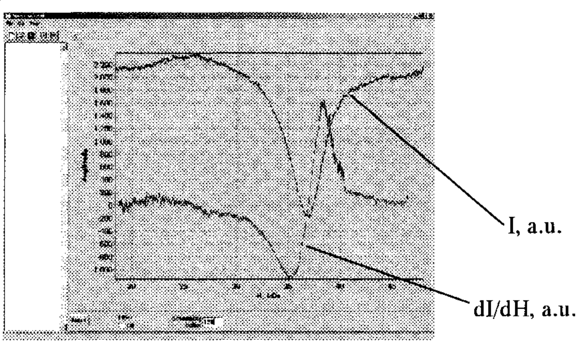

***Fig.2**. Typical FMR line shape for the CoFeMgF granular nanostructure layered specimen, (f=75.3 GHz, T=4.2K)*

As one can see the line has typically Lorentz shape with the width $\Delta H_{1/2}$=3-5 kOe both for T=300K and T=4.2K. This suggests the presence of strong exchange interactions, which are convenient for collinear ferromagnets. Let note that the width of the FMR line is narrower for the specimens with higher GMI/GMR response, (see Table 1 col.2-4).

It is a known fact that the line becomes narrower as the magnetic structure becomes more homogeneous. This experimental fact confirms the suggestion that GMI effect should be larger for more regular (crystal) structures then for amorphous ones.

Now let's analyze the resonance frequency-resonance field dependence ($\nu_{res} = f(H_{res})$) presented on Fig.3 for all of specimens ##1-4.

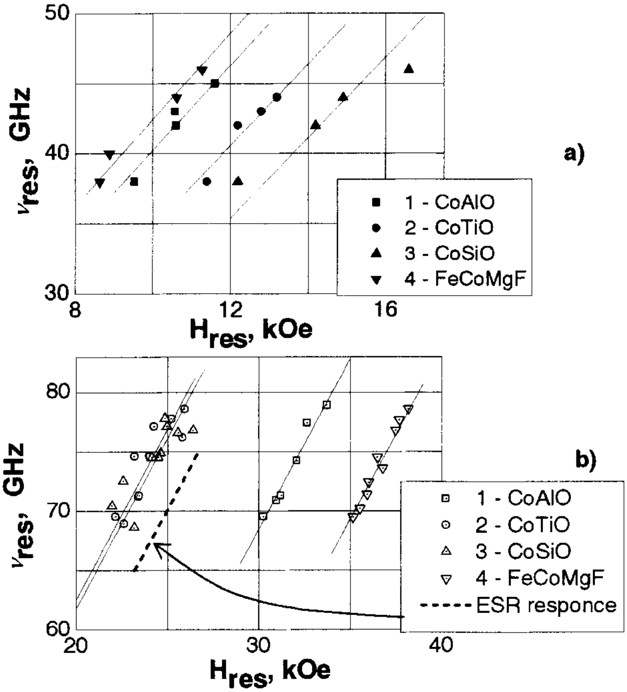

Fig.3. FMR experiment data for ##1-4 specimens:
a) the resonance frequency-field dependence for ||-orientation (T=300 K); b) the resonance frequency-field dependence for ⊥-orientation (T=4.2 K).

One can determine the value of the magnetization saturation (M_S) from these graphs by the known Kittel formulas

-for ||-orientation, i.e. vector H lies normally to the specimen plane surface (see inserts Fig.1.):

$$\nu_{res} = \frac{g\beta}{h}\sqrt{H_{res}(H_{res} + 4\pi M_S)} \quad (3),$$

and for ⊥-orientation, i.e. vector H lies in plane of the specimen:

$$\nu_{res} = \frac{g\beta}{h}(H_{res} - 4\pi M_S), \qquad (4),$$

where h is the Planck's constant, β is the Bohr magneton, g is the spectroscopic splitting factor.

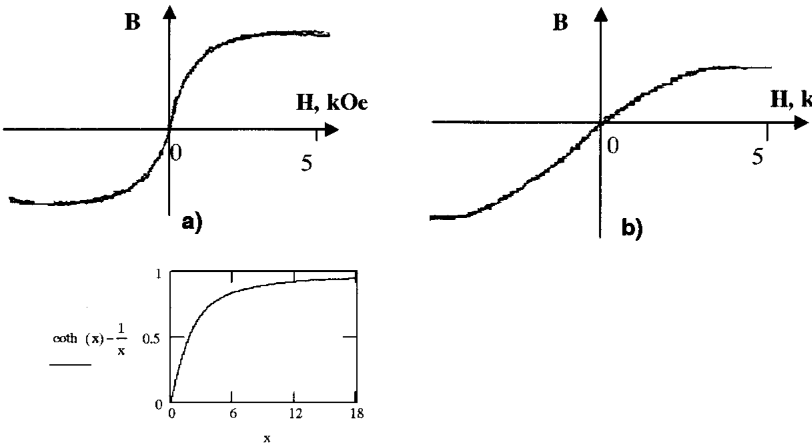

Fig.4. *Static magnetization for the CoSiO system at T=300K: a)- parallel orientation, b) –perpendicular orientation.*

The M_S magnitudes were extracted from data Fig.3a. It was suggested here that g-factor g=2.05±0.5%, that is true for the majority of ferromagnetics. This M_S magnitudes were confirmed as well by measuring $\nu_{res} = f(H_{res})$ dependence for perpendicular orientation and direct determination of g-value as a slope of linear function (4). The M_S data for T=300 K are presented in Table 1 col.5.

Dynamical data were compared with quasistatical ones. The main results of static measurements are the following. In-plane hysteresis loops (|| -orientation) have the form similar to Langevin function with very small hysteresis. Close approximation by this function (see insert in Fig.4) is possible for CoSiO and CoTiO systems. This fact is evidence of superparamagnetic behaviour of particles at present experiment conditions. For CoAlO and FeCoMgO systems under study some deviation from Langevin function were observed. Most likely it is connected with availability of some quantity of ferromagnetic phase. For all systems the mean magnetization (per unit of film volume) M_{S1} was calculated (Table 1 col.7).

The hysteresis loops measured normal to film plane ($\perp$-orientation) have the form shown in Fig. 4b and differ for investigated systems by the value of saturation field H_S. Using these data and known formula $H_S = NM_S$ (N is the demagnetizing term) a rough estimation of magnetization (per unit of particle volume) can be performed for the given systems composed of non-interacting particles. Comparison of parameters of ||- and $\perp$-orientations hysteresis loops allows supposing that nanogranulars are not correct spheres but some flattened ellipsoid. Therefore value of N is between $4\pi/3$ (spherical particle) and 4π (flat layer. Results of magnetization calculations (M_{S2}) for these outermost cases

are given in Table 1 col.7. The data of M_s measured by the dynamic (FMR) method (Table 1 col.6) as well lie in the interval of these data.
Thus it possible to conclude from this that for the real structures while the FMR-measurements it is necessary to take into account that magnetic granules have not spherical but ellipsoidal, flatted in the film plane, shape. The proportion extracted from the data in col. 5 and col. 7 should define the ratio between the ellipsoid axes

Table 1 (T=300 K)

1	2	3	4	5	6	7
specimen	$\Delta T/T$(%) ν=44 GHz δH= ±1.5кOe	$\Delta R/R$(%) δH= ±1.5кOe	$\Delta H_{1/2}$ (kOe)	M_S(G) (from FMR method)	M_{S1}(G) (from static method)	M_{S2}(G) (from static method)
1-CoAlO	2.28	5.08	3.58	768.8	310	440-1300
2-CoTiO	1.6	2.42	4.0	369.8	180	260-790
3-CoSiO	<<0.001	2.99	4.5	307.9	260	320-960
4-FeCoMgF	<<0.001	1.32	4.5	944.3	240	240-720

But the results that are of most interest were obtained in 4-mm waveband for the low temperatures (T=4.2 K). They are presented in Fig. 3.b.. Firstly, the unexpectedly large dispersion of experimental points for specimens ##2, 3 is observed. This dispersion exceeds in 1.5 orders the accuracy of experiment.
Secondly, for the same ##2, 3 samples the dependence $\nu_{res} = f(H_{res})$ lie on the left from the reference ESR dependence (see the dashed line $\nu_{res} = f(H_{res})$ for the ESR signal, i.e.: $M_S = 0$, $g = 2.00$). Applying the known formula (4) to calculate M_S one shall obtain the negative magnitude of magnetization of the specimen. This result is in contradiction with the physical reality.
The reasonable way to avoid this contradiction is to suggest that at cryogenic temperatures ($T \approx 4.2$ K) in specimens ##2, 3 some internal field appears (is induced). This internal field (let call it an effective anisotropy field H_{an}^{eff}) is directed along with the external field and appears only for ⊥-orientation.
The effective anisotropy field caused by the magnetostriction, which presents in the nanostructures, could produce such situation. Thin-layered nanostructures under study consist from spherical-like shaped-like nanogranules. Under the affect of large external magnetic field these spherical granules can change their shape to ellipsoidal one. Let's suggest that the large axis of ellipse is oriented perpendicular to the external field (H) direction (because the magnetostriction in these structures [7] is negative - $\lambda_S<0$). Thus a mechanical stress in the nanolayer appears. As a result of this stress the anisot ropy field, which is directed along the external H-field, appears.
However another explanation of the results observed is reasonable as well. The effect could be connected with the thermoelastic stress appearing at sample cooling since Co

has nonzero magnetostriction constant (λ_S<0) and with the fact that thermal-expansion coefficients of Co, glass substrate and dielectric matrix have different values. But estimations of such anisotropy field give the values not exceeding 1000 Oe. At the same time the calculation on the basis of FMR data lead to value ~ 5000-6000 Oe.

In spite of both of these suggestions are consistent each other, the question about origin of magnetic anisotropy revealed at low temperature in films under study is open now and will be studied especially.

Any case, for both of these suggestions, as the nanostructure is 2D system, so the anisotropy field (H_{an}^{eff}) appears to be:

- large if H is perpendicular to the specimen plane; - small if H is parallel to the specimen plane.

In this suggestion the Kittel formulas (3-4) for the resonance frequency–field dependence will be as following:

for $\|$-orientation
$$\nu_{res} = \frac{g\beta}{h}\sqrt{H_{res}\left(H_{res} + 4\pi M_S\right)} \qquad (5),$$

and for $\perp$-orientation
$$\nu_{res} = \frac{g\beta}{h}(H_{res} - 4\pi M_S + H_{an}^{eff}) \qquad (6),$$

Accepting this suggestion we have calculated the magnitude of H_{an}^{eff} from millimeter waveband experiment for T=4.2 K.

Table 2 (T=4.2 K)

1	2	3	4
specimen	$S = 4\pi M - H_{an}^{eff}$ (Oe)	H_{an}^{eff} (Oe)	M_S (G)
1-CoAlO	6154	0	490
2-CoTiO	-1871.8	5891	320
3-CoSiO	-1249.7	6864	447
4-FeCoMgF	10863	0	865

Experimental data were processed by the following way:

1. Data from Fig.3.b were approximated by the modified Kittel equation (6);
2. From (6) and the Fig.3.b we extract directly the g=2.05 as a slope of linear dependence $\nu_{res} = f(H_{res})$. Then we find the sums $S=4\pi M - H_{an}^{eff}$, which are shown in Table 2 (col 2).
3. While solving the system (5-6) we obtain the magnetization saturation (M_S) (Table 2, col 4) and the anisotropy field (H_{an}^{eff}) (Table 2 col.3) magnitudes for specimens ##2, 3;
4. Following the results arise from Fig.3.b to define the magnetization saturation for ##1, 4 we use the same equation (6), with the assumption that $H_{an}^{eff} = 0$. So for these samples we obtain from (6) M_S magnitudes directly (Table 2 col.4).

Thus using the assumption about the anisotropy field we obtain quite reasonable magnitudes of the magnetization for T=4.2K (Table 2), which are not contradict with data for T=300 K (Table 1).

4. Conclusions

Thus we can summarize the outcomes of the millimeter waveband FMR studies of GMI nanostructures.

1. The FMR response for 37-80 GHz at T=300 K and T=4.2 K was detected the millimeter waveband. Noticeable GMI magnitudes in millimeter waveband (f=44GHz) for specimens ##1, 2 were detected.
2. The linewidth magnitude $\Delta H_{1/2}$ varies between 3-5 kOe. The linewidth correlates with the GMI magnitude in the millimeter waveband that proves the suggestion that GMI is larger for magnetically homogenous nanostructures.
3. Magnetic nanostrucures under study demonstrate the superparamagnetic behavior at room temperatures.
4. The presence of perpendicular anisotropy is suggested at cryogenic temperatures. The magnitudes of the effective anisotropy field and saturation magnetization were estimated.

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