JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. **5** No 4, 04075(5pp) (2013)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том **5** № 4, 04075(5cc) (2013)

Effect of Indirect Exchange Interaction on Ferromagnetic Resonance in Magnetic Nanogranular Films

Yu.I. Dzhezherya, A.F. Kravets, I.M. Kozak, E.V. Shypil, A.M. Pogorily

Institute of magnetism of NASU, 36-b, Vernadsky Pr., 03142 Kyiv, Ukraine

(Received 22 November 2013; published online 31 January 2014)

The effect of the non-magnetic matrix conducting properties on the ferromagnetic resonance of the ensembles of ferromagnetic metal nanogranules in magnetic nanogranular films with different types of non-magnetic matrix (metal and dielectric) was investigated. Complex investigations of the behavior of nanogranular systems in ferromagnetic resonance showed that in materials with conducting matrix the evidence of ferromagnetic ordering occurs well before the structural percolation threshold. The experimental results are explained in terms of the phenomenological approach based on the consideration of the exchange bias of conduction electrons by magnetic ions of nanogranules. It is shown that the polarization in the subsystem of conduction electrons contributes to the appearance of the exchange interaction between the magnetic moments of granules.

Keywords: Nanogranular films, Magnetic granule, Ferromagnetic resonance, Exchange coupling, Structural percolation.

PACS numbers: 76.50. + g, 76.20. + q, 75.75. + a, 71.70.Gm, 61.46. – w, 45.70. – n

1. INTRODUCTION

Interest to magnetic nanogranular materials containing nanogranules of metallic ferromagnetic in non-magnetic matrix is conditioned by striking physical effects which are manifested in these systems under the magnetic field action. Giant magnetoresistance in the films with metallic magnetic and non-magnetic components [1-5], tunnel magnetoresistance in the metal-dielectric films [6-10], giant Hall effect [11], magnetorefractive effect [12-15], magnetically-induced nonlinear optical effects [16], etc. are among the mentioned effects.

Depending on the conducting properties of non-magnetic matrix, nanogranular films are divided into two basic classes – metal and metal-dielectric.

If distinctions in the transport properties of granular films with different matrix types are evident [17, 18], then interpretation of distinctions in the behavior of such systems during magnetization and ferromagnetic resonance (FMR) causes difficulties. From the point of view of the magnetic properties, both conducting and dielectric matrices are scarcely different, since their magnetic permeabilities are close to unit.

Analysis of the results of the magnetic investigations allows to suppose that additional correlation mechanism exists between magnetic moments of nanogranules in materials with conducting matrix. It can be connected with indirect exchange through electrons of the conduction band.

In contrast, in nanogranular materials with dielectric matrix indirect exchange is characterized by a small quantity, since at concentrations of magnetic material below the structural percolation threshold it is conditioned by the effects of spin-dependent tunneling of electrons from granule to granule through high potential barrier [6-10].

The most known theory of indirect exchange interaction in conducting systems is the Ruderman-Kittel-Kasuya-Yosida (RKKY) theory [19-22]. However, its application in the description of magnetic interaction in ferromagnetic systems is limited by the requirements of absence of the long-range magnetic order and equality to zero of the average magnetization in the system of conduction electrons. Therefore, pure RKKY exchange is exhibited only in diluted alloys with rare-earth atoms. Role of the RKKY exchange interaction in ensemble of nanogranules is rather insignificant. Really, influence on a separate magnetic granule on the side of neighboring magnetic clusters will be extremely minor because of fast spatial oscillations of the exchange interaction potential of the RKKY theory [23, 24].

Another mechanism of indirect exchange which is not considered by the RKKY theory can be connected with polarization of subsystem of conduction electrons due to the *s-d* exchange in the system with long-range magnetic order.

Indeed, if magnetize the ensemble of magnetic nanogranules being initially in the superparamagnetic state by external field, then magnetic polarization of the conduction *s*-electrons will be realized in the whole volume of the material. Influence of the magneto-polarized medium filling up the whole space is identical to the effective magnetic field which promotes the establishment of magnetic ordering in ensemble of nanogranules [25].

In contrast to metal nanogranular films, collective behavior of the ensemble of nanogranules in metal-dielectric films becomes significant only when the structural percolation threshold is achieved.

The primal problem of the experimental part of the present work consisted in the detection of the role of indirect exchange interaction when the magnetic order in nanoranular films is established. Distinctions in the collective behavior of the ensembles of granules in the systems with non-magnetic matrices of different types are clearly traced in the experiments on magnetization and ferromagnetic resonance [26-29].

The phenomenological approach for the description of indirect exchange interaction in conducting nanogranular materials is proposed in the theoretical part of the work. Interpretation of the experimental results of the FMR is given there.

2. EFFECT OF THE MATRIX CONDUCTING PROPERTIES ON THE FMR IN MAGNETIC NANOGRANULAR FILMS

For the investigation of the effect of the conducting properties of non-magnetic matrices on the FMR, metal Py_xAg_{1-x} and metal-dielectric $Py_x(Al_2O_3)_{1-x}$ films on ceramic substrates with the same material of magnetic nanogranules (Py – permalloy ($Fe_{20}Ni_{80}$)) but with different non-magnetic matrices (Ag and Al_2O_3) were produced in nanogranular structures. Film thickness was equal to ~ 200 nm. Volume content x of permalloy was varied in the range of 0.13-0.83 for metal and 0.13-0.47 for metal-dielectric films. These films were manufactured in the same conditions by simultaneous condensation on the substrates of magnetic (Py) and non-magnetic (Ag or Al_2O_3) components of granular alloys simultaneously evaporated in vacuum by the electron-beam method in accordance with the technique given in [30]. Details of the production of nanogranular films are represented in the work [31]. Ensembles of nanogranules in the films of both series below the structural percolation threshold ($x < 0.33$) are characterized by some distribution over the sizes within the range of $1 \div 5$ nm [32].

FMR spectra were obtained for films with different content of the magnetic component (Fig. 1 and Fig. 2). Information about the character of magnetic interaction in granular films was intended to obtain from the analysis of the concentration dependences of resonance fields defined at the parallel and perpendicular orientations of the film plane in magnetic field (Fig. 3).

The Kittel formulas denote that the lower average magnetization of the longitudinal and transverse resonance magnetic fields in ferromagnetic films is, the less differences in these fields are [33]. Therefore, as it follows from Fig. 3, at small concentrations of magnetic inclusions magnetization of granular structures with any matrix type is extremely insignificant. It may be assumed that magnetic granules are in the superparamagnetic state. With the increase in the concentration of ferromagnetic material conducting properties of the matrix qualitatively impact on the magnetic state of granular films. Thus, if in metal-dielectrics the superparamagnetic state continues up to the structural percolation threshold ($x \approx 0.3$), then in metal nanogranular systems the additional correlation mechanism joins well before (at $x > 0.18$). At concentrations of magnetic material of $0.18 < x < 0.3$ structural percolation of granules in metal films does not occur yet, but signs of the magnetic ordering appear.

For the explanation of the discrepancies in the concentration dependences of the FMR resonance field for systems with different types of matrix conduction, one can use different physical models.

In particular, the model used in the present work is based on the introduction of the concept of an effective field. Thus, during magnetization of the ensemble of nanogranules by an external field in the system of free s-electrons, magnetic polarization appears due to the interband interaction with d-electrons of magnetic nanogranules. Effect of the magnetically polarized medium is described by some effective field, which promotes the establishment of magnetic order prior to the structural percolation threshold.

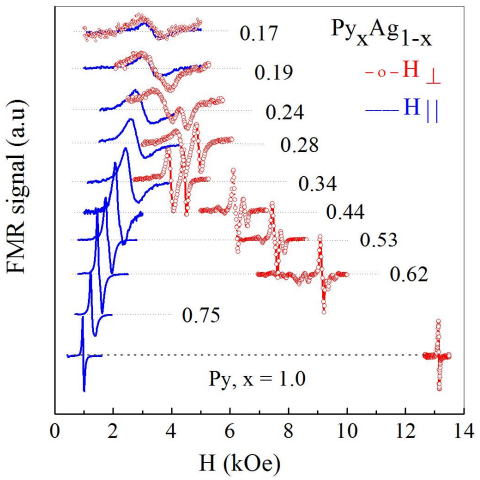

Fig. 1 – FMR spectra for metal Py_xAg_{1-x} nanogranular films of different contents for the parallel (lines) and perpendicular (circles) orientations of the film plane with respect to the direction of the applied magnetic field

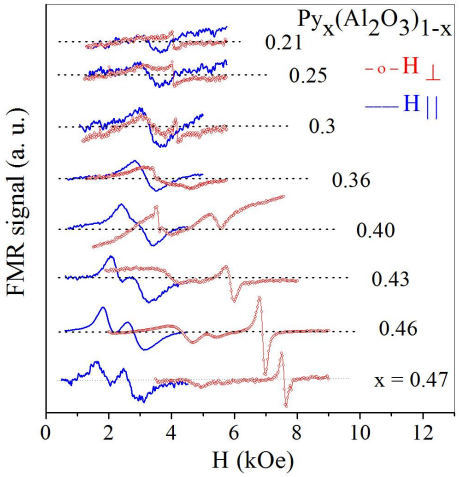

Fig. 2 – FMR spectra for metal-dielectric $Py_x(Al_2O_3)_{1-x}$ nanogranular films of different contents for the parallel (lines) and perpendicular (circles) orientations of the film plane with respect to the direction of the applied magnetic field

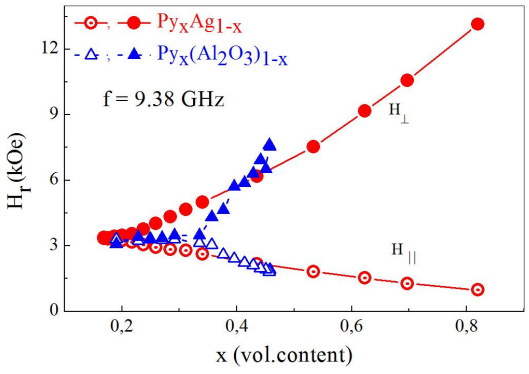

Fig. 3 – Concentration dependences of the FMR resonance fields for metal Py_xAg_{1-x} and metal-dielectric $Py_x(Al_2O_3)_{1-x}$ films for the perpendicular (●, ▲) and parallel (○, Δ) film orientations in magnetic field

The theory which qualitatively describes the dependences of the resonance fields was proposed in the work [27]. However, it was assumed within this work that

system magnetization is close to saturation. Evidently, such assumption is true at large concentrations of magnetic inclusions. At the same time, at small concentrations of magnetic material in conditions of room temperature ensemble of nanogranules is in the superparamagnetic state. Therefore, concentration dependences of the resonance fields calculated in [27] slightly differ from the experimental data.

Let us construct the phenomenological model for the description of the features of the concentration dependences of the FMR resonance fields.

For simplification, we will suggest that granules of ferromagnetic have spherical shape and are characterized by a weak crystalline anisotropy.

In this case, magnetic moments of granules are under the action of the effective field

$$\mathbf{H}^{eff} = \mathbf{H} - 4\pi M_z \mathbf{e}_z + \mathbf{h}\,, \qquad (1)$$

where **H** is the external magnetic field; the second term determines the film demagnetizing field; term **h** is introduced nominally and describes the indirect exchange field through the electron subsystem.

For simplification, spread of the local fields connected with the irregularity of the distribution of ferromagnetic nanogranules in the film volume was not taken into account in (1).

Using the results of the work [25], in which the phenomenological theory allowing to define the value of the indirect exchange effective field was developed, the value of **h** can be represented in the form

$$\mathbf{h} = J^2 \chi \mathbf{M} = k\mathbf{M}\,, \qquad (2)$$

where J is the constant of the interband s-d exchange interaction; χ is the paramagnetic susceptibility of the subsystem of conduction electrons.

For a more detailed acquaintance with formula (2) derivation and its application conditions, one can appeal to the original work [25]; here we will be limited by the introduction of the phenomenological parameter $\kappa = J^2\chi$, whose value is always positive.

Dependence of the FMR frequency on the external field is determined from the Landau-Lifshitz equation for the dynamics of the average system magnetization obtained as a result of averaging of the motion equations of the magnetic moments of separate granules.

$$\frac{d\mathbf{M}}{dt} = \gamma\left[\mathbf{M}\times\mathbf{H}^{\text{eff}}\right],\ \gamma = g\mu_{\text{B}}/\hbar\,, \qquad (3)$$

where $\mathbf{M} = (1/V)\sum_i \mathbf{m}_i$ is the vector of the average system magnetization; V is the system volume; $\mathbf{m}_i$ is the magnetic moment of the i-th granule; g is the gyromagnetic ratio; μ_{B} is the Bohr magneton; $\hbar$ is the Plank constant.

If suppose that system is located in the uniform magnetic field and consider small oscillations of magnetization in the vicinity of the ground state, then FMR frequency will be defined by the Kittel formulas [33]

$$\omega_{\text{II}} = \gamma\sqrt{H_{\text{II}}(H_{\text{II}} + 4\pi M_{\text{II}})}\,. \qquad (4a)$$

If magnetic field $H_\perp$ is applied perpendicularly to the film plane, then FMR frequency is determined from the following relation:

$$\omega_\perp = \gamma(H_\perp - 4\pi M_\perp)\,. \qquad (4b)$$

The values of M_{II} and $M_\perp$ denote the average values of the planar and normal components of the system magnetization at the corresponding orientation of the external magnetic field.

At first glance, indirect exchange interaction has no effect on the dependences of resonance frequencies (4a) and (4b). However, the presence of this interaction can considerably influence the value of the average system magnetization.

Indeed, assuming that magnetic moments of granules are in the superparamagnetic state, we will apply the Langevin approach and write that average magnetization of the ensemble of granules is equal to

$$M = \frac{1}{V}\sum_i \langle m_i\rangle = \frac{1}{V}\sum_i m_i L(m_i H^{\text{eff}}/k_{\text{B}}T)\,, \qquad (5)$$

where $L(z) = \cot g(z) - 1/z$ is the Langevin function; k_{B} is the Boltzmann constant.

For simplification of the calculations we will use the model approximation considering ensemble of granules of the same size. In this case, correlation (5) will be rewritten as follows:

$$M = x\cdot M_0 L(\upsilon M_0 H^{\text{eff}}/k_{\text{B}}T)\,, \qquad (6)$$

where x is the volume concentration of ferromagnetic granules; M_0 is the saturation magnetization of the granule material; υ is the granule volume.

Thus, using expression (6) and formulas (4a) and (4b) for FMR frequencies, we obtain the correlations which define the dependence of the FMR resonance magnetic field on the concentration of ferromagnetic material in granular film.

Thus, for a film in the planar field we have

$$1 - h_{\text{II}}^2 - x\cdot(4\pi M_0/H_\omega)\cdot h_{\text{II}} L\left(\frac{\Theta}{T}\left(h_{\text{II}} + \frac{\kappa}{4\pi}\frac{1-h_{\text{II}}^2}{h_{\text{II}}}\right)\right) = 0\,; \qquad (7a)$$

for a film in the perpendicular film

$$h_\perp - 1 + x\cdot(4\pi M_0/H_\omega)\cdot L\left(\frac{\Theta}{T}\left(1 + \frac{\kappa}{4\pi}(h_\perp - 1)\right)\right) = 0\,, \qquad (7b)$$

where designations $H_\omega = \omega/\gamma$, $h_\perp = H_\perp/H_\omega$, $h_{\text{II}} = H_{\text{II}}/H_\omega$, $\Theta = \upsilon\cdot M_0 H_\omega/k_{\text{B}}$ are introduced.

Two phenomenological parameters Θ and κ enter into the relations (7a) and (7b). In turn, parameter Θ depends on the value of ν, which in the sense corresponds to the characteristic volume of granules.

Correspondence between the theoretical and experimental dependences of the resonance fields on the concentration of magnetic material in metal nanogranular Py_xAg_{1-x} films is achieved by the fitting of the values Θ and κ (Fig. 4). The best coincidence of the experimental and theoretical dependences of the resonance fields on

the volume content of ferromagnetic material is achieved at the following parameters: $T/\Theta \approx 8$ and $\kappa/4\pi \approx 26$ that corresponds to $M_0 \approx 800$ Gs, $v \approx 1.5 \cdot 10^{-21}$ cm^3. The value of saturation magnetization M_0 of separate nanogranules, which is used in the calculations, is much less than in uniform ferromagnetic. The fact of the decrease in the saturation magnetization of nanogranules with the decrease in their size was discussed in [34].

We note that correlations (7a) and (7b) are true only at small concentrations of magnetic material (below the structural percolation threshold).

In contrast to the systems with conducting matrix, collective effects in metal-dielectric nanogranular materials become apparent only when the structural percolation threshold is reached. It is shown in Fig. 2 and Fig. 3 that for the set of $Py_x(Al_2O_3)_{1-x}$ samples one can observe the phase transition at concentrations of ferromagnetic material close to $x_p \approx 0.33$. Obviously, ferromagnetic complexes, whose magnetic moments rather easily orient along the applied field, appear at x_p. With further increase in the concentration of x fraction of nanogranules in the superparamagnetic state decreases, and fraction of ferromagnetic complexes increases.

Correlations (7a) and (7b) are also true for metal-dielectric films before the percolation threshold $x < x_p$, with the difference that for them the constant of indirect exchange should be set to zero ($\kappa = 0$). In Fig. 4 we compare the theoretical and experimental concentration dependences of the FMR resonance fields for metal Py_xAg_{1-x} and metal-dielectric $Py_x(Al_2O_3)_{1-x}$ nanogranular films. Theoretical calculations are performed on the basis of formulas (7a) and (7b).

Analysis of the results shows that indirect exchange interaction in the nanogranular system with conducting matrix promotes the establishment of magnetic correlation well before the structural percolation threshold.

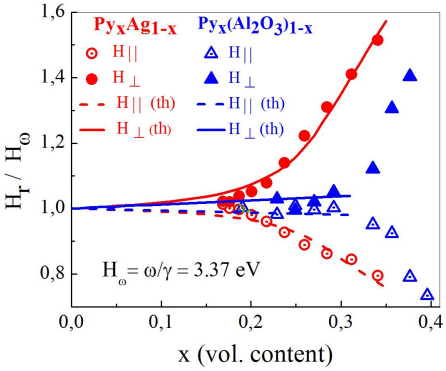

Fig. 4 – Comparison of the calculation (lines) and experimental (dots) concentration dependences of the FMR resonance fields for metal Py_xAg_{1-x} and metal-dielectric $Py_x(Al_2O_3)_{1-x}$ films for the perpendicular (the upper branch of lines) and parallel (the lower branch of lines) film orientations in magnetic field

It follows from the estimations carried out in the present work that $\kappa/4\pi \approx 26$. On the other hand, it is shown in the work [25] that $\kappa = J^2\chi$(where J is the constant of the interband s-d exchange interaction; χ is the paramagnetic susceptibility of the subsystem of conduction electrons). Hence it appears that the value of constant is equal to $J \approx 5 \cdot 10^3$ for typical values $\chi \approx 10^{-5}$.

As for the radius of exchange interaction, it can be determined from the concentration dependences of the resonance fields. Exchange interaction in granular materials with metal matrix is manifested at the volume concentration of magnetic component $x_c \approx 0.18$. In this case, $V = v/x$ (where v is the granule volume; x is the volume concentration of magnetic material) is the volume per one granule. It follows that average distance between centers of neighboring granules will be equal to $V^{1/3}$. Thus, distance R between centers of granules can be estimated by the formula $R \approx d/x_c^{1/3}$ (where d is the average diameter of a granule).

Taking into consideration that average diameter of magnetic granules in nanogranular films at concentrations $x_c \approx 0.18$ is equal to 1.2 nm [32], then $R \approx 2$ nm and distance between the surfaces of neighboring granules is about 0.8-1.0 nm.

Thus, exchange interaction between granules is characterized by high intensity but short radius of action.

3. CONCLUSIONS

Effect of the matrix conducting properties of nanogranular systems on the collective properties is clearly traced in the experiments on ferromagnetic resonance. It is revealed that indirect exchange interaction in the nanogranular system with conducting matrix Py_xAg_{1-x} promotes the establishment of magnetic correlation and magnetic ordering in the system of magnetic nanogranules well before the structural percolation threshold. Exchange interaction in Py_xAg_{1-x} films starts appear for the volume concentration of magnetic component $x_c \approx 0.18$.

In contrast to the systems with conducting matrix, collective effects in metal-dielectric nanogranular materials $Py_x(Al_2O_3)_{1-x}$ become apparent only when the structural percolation threshold is reached at the concentrations of ferromagnetic material close to $x_p \approx 0.33$.

Exchange interaction between granules is characterized by high intensity and short radius of action.

AKNOWLEDGEMENTS

The authors of the paper express their gratitude to A.Ya. Vovk for the determination of the content of investigated films. The work has been performed under the partial financial support of the grant of international cooperation NSF # 9940368.

REFERENCES

1. A.E. Berkowitz, J.R. Mitchell, M.J. Carey, A.P. Young, S. Zhang, F.E. Spada, F.T. Parker, A. Hutten, G. Thomas, *Phys. Rev. Lett.* **68**, 3745 (1992).
2. J.Q. Xiao, J.S. Jiang, C.L. Chien, *Phys. Rev. Lett.* **68**, 3749 (1992).
3. C.L. Chien, J.Q. Xiao, J.S. Jiang, *J. Appl. Phys.* **73**, 5309 (1993).
4. A. Gerber, A. Milner, B. Groisman, M. Karpovsky, A. Gladkih, A. Sulpice, *Phys. Rev. B* **55**, 6446 (1997).
5. S. Honda, M. Nawate, M. Tanaka, T. Okada, *J. Appl. Phys.* **82**, 764 (1997).
6. S. Honda, T. Okada, M. Nawate, M. Tokumoto, *Phys. Rev. B* **56**, 14566 (1997).
7. S. Mitani, S. Takahashi, K. Takanashi, K. Yakushiji,

S. Maekawa, H. Fujimori, *Phys. Rev. Lett.* **81**, 2799 (1998).
8. S. Sankar, A.E. Berkowitz, D. Smith, *Phys. Rev. B* **62**, 14273 (2000).
9. K. Yakushiji, S. Mitani, K. Takanashi, S. Takahashi, S. Maekawa, H. Imamura, H. Fujimori, *Appl. Phys. Lett.* **78**, 515 (2001).
10. A.Ya. Vovk, J.-Q. Wang, W. Zhou, J. He, A.M. Pogoriliy, O.V. Shypil, A.F. Kravets, H.R. Khan, *J. Appl. Phys.* **91**, 10017 (2002).
11. A.B. Pakhomov, X. Yan, B. Zhao, *J. Appl. Phys.* **79**, 6140 (1996).
12. A.B. Granovski, M.V. Kuzmichev, J.P. Clerk, *J. Exp. Theor. Phys.* **89**, 955 (1999).
13. V.G. Kravets, D. Bozec, J.A.D. Matthew, S.M. Thompson, H. Menard, A.B. Horn, A.F. Kravets, *Phys. Rev. B* **65**, 054415 (2002).
14. A.B. Granovski, I.V. Bykov, E.A. Gan'shina, V.S. Gushchin, M. Inoue, Yu.E. Kalinin, A.A. Kozlov, A.N. Yurasov, *J. Exp. Theor. Phys.* **96**, 1104 (2003).
15. V.G. Kravets, L.V. Poperenko, I.V. Yurgelevich, A.M. Pogorily, A.F. Kravets, *J. Appl. Phys.* **98**, 043705 (2005).
16. O.A. Aktsipetrov, T.V. Murzina, E.M. Kim, R.V. Kapra, A.A. Fedyanin, M. Inoue, A.F. Kravets, S.V. Kuznetsova, M.V. Ivanchenko, V.G. Lifshits, *J. Opt. Soc. Am. B* **22**, 138 (2005).
17. S. Zhang, P.M. Levy, *J. Appl. Phys.* **73**, 5315 (1993).
18. J. Inoue, S. Maekawa, *Phys. Rev. B* **53**, R11927 (1996).
19. M.A. Ruderman, C. Kittel, *Phys. Rev.* **96**, 99 (1954).
20. T. Kasuya, *Progr. Theor. Phys.* **16**, 45 (1956).
21. K. Yosida, *Phys. Rev.* **106**, 893 (1957).
22. C. Kittel, *Quantum Theory of Solids* (Wiley, New York: 1963).
23. G.M. Genkin, M.V. Sapozhnikov, *Appl. Phys. Lett.* **64**, 794 (1994).
24. P. Vargas, D. Altbir, *J. Magn. Magn. Mater.* **167**, 161 (1997).
25. Yu.I. Gorobets, Yu.I. Dzhezherya, A.F. Kravets, *Phys. Solid State* **42**, 126 (2000).
26. B. Dieny, S.R. Teixeira, B. Rodmacq, C. Cowache, S. Auffer, O. Redon, J. Pierre, *J. Magn. Magn. Mater.* **130**, 197 (1994).
27. D. Pod'yalovskii, A. Pohorilyi, B. Ivanov, A. Kravets, C.S. Kim, *J. Magn. Magn. Mater.* **196-197**, 131 (1999).
28. G.N. Kakazei, A.F. Kravets, N.A. Lesnik, M.M. Pereira de Azevedo, Yu.G. Pogorelov, J.B. Sousa, *J. Appl. Phys.* **85**, 5654 (1999).
29. Yu.G. Pogorelov, G.N. Kakazey, J.B. Sousa, A.F. Kravets, N.A. Lesnik, M.M. Pereira de Azevedo, M. Malinowska, P. Panissod, *Phys. Rev. B* **60**, 12200 (1999).
30. G.A. Niklasson, C.G. Grandvist, *J. Appl. Phys.* **55**, 3382 (1984).
31. N. Pohorilyi, A.F. Kravets, E.V. Shypil', D.Y. Pod'yalovsky, A.Ya. Vovk, C.S. Kim, M.V. Prudnikova, H.R. Khan, *Thin Solid Films* **423**, 218 (2003).
32. C.J. O'Connor, V.O. Golub, A.Ya. Vovk, A.F. Kravets, A.M. Pogoriliy, *IEEE T. Magn.* **38**, 2844 (2002).
33. C. Kittel, *Phys. Rev.* **73**, 155 (1948).
34. S.N. Khanna, S. Linderoth, *Phys. Rev. Lett.* **67**, 742 (1991).