JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 10, No. 10, October 2008, p. 2678 - 2683

High frequency simulation of granular CoFeHfO nanometric thin films

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The granular CoFeHfO material with good soft magnetic properties has been analyzed. This magnetic material is used like thin films with nano-metric thickness, deposed by reactive sputtering on a printed circuit board substrate surface planarized beforehand by chemical-mechanical polishing. Our study is performed in order to determine the electric and magnetic properties of efficient package-compatible magnetic materials, which are needed for exploitation of magnetic passive components and devices. Two important material parameters have been determined: relative effective electric permittivity and relative magnetic permeability, using a simulation method in the frequency range of 2.45 - 9.8 GHz. The material parameters have been calculated according to an algorithm based on physical considerations, applied for the material samples exposed to a testing electromagnetic field. Our new contribution is to illustrate the resonant character of the frequency behavior for the material parameters (the effective electric permittivity and the magnetic permeability). This resonant behavior is connected with the microscopic internal structures of the samples, depending on its geometry, internal order and nature.

(Received November 12, 2007; accepted August 14, 2008)

Keywords: Granular CoFeHfO, Nano-metric thin films, Relative effective electric permittivity and relative magnetic permeability, Physical algorithm, Resonant high-frequency behavior, Internal order.

1. Introduction

The amorphous cobalt alloy CoFeHfO is used like magnetic nano-metric thin films for high-frequency magnetic device applications. The material is magnetic and electric anisotropic and present specific properties which make it an efficient package-compatible magnetic material. Thin films of CoFeHfO are used to manufacture the high-frequency magnetic devices, such as thin-film inductors and transformers for microswitching converters and ultrahigh-density recording heads [3]. The CoFeHfO thin films present low damping constant, in conjunction with the high magnetic anisotropy, large resistivity, large high-frequency susceptibility (in GHz domain), and large spin-dependent-tunneling magnetoresistance. In addition, films show a weak dependence of the film resistivity, high-frequency damping, and magnetic properties on temperature in the range between 4 K and 300 K [4], [7].

Material composition can be described using the formula: $(Co_{1-c}\ Fe_c)_x\ Hf_y\ O_z$, $0.05 \le c \le 0.5$; $3 \le y \le 30$, $7 \le z \le 40$; x is the balance (the alloy also containing inevitable impurities). The role of each element is as follows: Co and Fe are main components, and Co, Fe and Ni are elements which bear magnetism; the contents of Co and Fe are preferably as high as possible. Hf is necessary for obtaining soft magnetic characteristics and combines with oxygen to form an oxide; Hf is considered as having the function to suppress magnetostriction [4], [13]. The internal order of the material is interesting, too, because we have here a multi-phases structure. Fe stabilizes the face-centered cubic structure (fcc structure) of Co or increases uniaxial anisotropy (Co addition to Fe-Hf-O films improves the frequency characteristics mainly by the increase in the crystalline anisotropy of the nanograins). In the texture of the soft magnetic alloy film, a microcrystalline phase may have a body-centered cubic structure (bcc structure), and the texture may be a mixed texture comprising a microcrystalline phase of the fcc structure, a microcrystalline phase of the bcc structure and the remainder mainly comprising an amorphous phase containing Co and element Fe. The alloy film with this complex texture and composition has uniaxial anisotropy within a crystal face [2], [14].

Material nano-films are fabricated by sputtering, vapor deposition or the like. There are used sputtering apparatus such as: a RF double-pole sputtering, DC sputtering, magnetron sputtering, triple-pole sputtering, ion beam sputtering or opposite target type sputtering apparatus. After film deposition, in order to improve the soft magnetic characteristics of the film, the film was annealed by maintaining it at a temperature within the range of 300° to 600° C for 60 to 360 minutes in a static magnetic field, and then slowly cooling the film [2], [3], [8].

For a high-frequency properties study it is also important that the CoFeHfO layers have a high in-plane induced anisotropy field (tens of Oe) and a high M_s value (of 1T order), leading to a ferromagnetic resonance frequency higher than a few GHz [2], [7], [9].

Material simulation methods are used for electro-magnetic properties determination at high frequencies. These techniques assist the electronic devices design process. A typical simulation might divide the device into small portions and approximate each with a cell of uniform magnetization; the interactions between these cells then describe the behavior of the whole material [12]. Other simulation methods, like the complex GRAPE (*GrAvity PipE)* technique consider the continuous magnetic material like an array of discrete dipoles that could respond individually to the magnetic fields in their neighborhoods (the method involves many hard resources) [11]. We have worked here developing a structure simulation strategy, in which the geometrical components of the material samples were re-built, at microscopic level, and their geometrical and physical characteristics were attached properly. The simulated magnetic material was then exposed to a testing HF electromagnetic field and the filed modifications were detected after the interaction with material sample [10], [11], [12].

The physical algorithm for material parameters determination was described in the considerations bellow.

2. Material parameters determination

The material parameters like the electric effective permittivity and the magnetic permeability were determined here, for granular the CoFeHfO nano-metric thin films. Simulations were done by computer implementation of a physical algorithm, which consider the material samples exposed to a testing field in the HF range (microwave domain). The electric and magnetic anisotropy of the material requires a tensor description of the material parameters (electric permittivity and magnetic permeability), illustrated in the relations bellow[1]:

$$\boldsymbol{P} = \varepsilon_0 [\chi_e] \boldsymbol{E} = \varepsilon_0 ([\varepsilon_r] - [1]) \boldsymbol{E} \tag{1}$$

$$\boldsymbol{M} = [\chi_m] \boldsymbol{H} = ([\mu_r] - [1]) \boldsymbol{H}\ , \tag{2}$$

where the $\boldsymbol{P}$, respectively $\boldsymbol{M}$ vectors are the material polarizability and the magnetization; $[\chi_e]$, respectively $[\chi_m]$ are the electric and magnetic susceptivity tensors; $[\varepsilon_r]$, respectively $[\mu_r]$ are the electric relative permittivity tensor and the magnetic relative permeability tensor and ε_0 is the electric permittivity of the free space.

The electric, respectively magnetic field energy variation at material sample polarization/magnetization can be written as [1]:

$$dw_e = \boldsymbol{E} \cdot d\boldsymbol{P} \tag{3}$$

$$dw_m = \mu_0 (\boldsymbol{H} \cdot d\boldsymbol{M}), \tag{4}$$

where w_e and w_m represent the volumetric densities of energy.

For the anisotropic material case, the relations above can be developed:

$$dw_e = \varepsilon_0 \cdot (\boldsymbol{E} \cdot [\chi_e] \cdot d\boldsymbol{E}) \tag{5}$$

$$dw_m = \mu_0 \cdot (\boldsymbol{H} \cdot [\chi_m] \cdot d\boldsymbol{H}), \tag{6}$$

which are equivalent with:

$$\begin{aligned} dw_e = dE_x &\left[(\varepsilon_{11} - 1)E_x + \varepsilon_{21} E_y + \varepsilon_{31} E_z\right] + \\ + dE_y &\left[\varepsilon_{12} E_x + (\varepsilon_{22} - 1)E_y + \varepsilon_{32} E_z\right] + \\ + dE_z &\left[\varepsilon_{13} E_x + \varepsilon_{23} E_y + (\varepsilon_{33} - 1)E_z\right] \end{aligned} \tag{7}$$

$$\begin{aligned} dw_m = dH_x &\left[(\mu_{11} - 1)H_x + \mu_{21} H_y + \mu_{31} H_z\right] + \\ + dH_y &\left[\mu_{12} H_x + (\mu_{22} - 1)H_y + \mu_{32} H_z\right] + \\ + dH_z &\left[\mu_{13} H_x + \mu_{23} H_y + (\mu_{33} - 1)H_z\right] \end{aligned} \tag{8}$$

When we set the proper testing field component and/or its variations to zero, the electric permittivity, respectively magnetic permeability components are determined. For example, to obtain ε_{12}, the components E_y, E_z and the variations dE_x and dE_z are to be vanished. The permittivity component will then be:

$$\varepsilon_{12} = \left.\frac{dw_e}{E_x \cdot dE_y}\right|_{\substack{E_y,\, E_z = 0 \\ dE_x,\, dE_z = 0}} \tag{9}$$

Variation of volumetric densities of energy inside the material, dw_e, respectively dw_m are given individually by the simulation program. They correspond to the exposure field strength, imposed by the user who sets the $\boldsymbol{E}$ and $\boldsymbol{H}$ field components values.

The method has the advantage that the testing field can be applied on an arbitrary direction in respect to the anisotropy axes of the material. In addition, the method facilitates determination of the permittivity and permeability tensor components, which are difficult to be obtained by other methods. The method also considers that the anisotropic material presents more than one constituents and the permittivity/permeability are effective ones, depending on constituents nature and geometry and also of the environment characteristics (tempetature, humidity, etc.).

3. Simulation results and interpretations

Simulations were performed for the granular CoFeHfO material, deposed like nano-metric thin films on a printed circuit board substrate (halogen-free FR-4). The films

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[1] the quantities bold written are 3D vectors.

thickness is of 10 nm (the average crystal grain size is of about 3-5 nm), on a substrate of 0.8 mm (Fig. 1). In practice, a thin film is deposed by reactive sputtering in a static magnetic field on the substrate surface, well planarized by chemical-mechanical polishing (a rough substrate surface degrades the magnetic property of the film). After film deposition, for improving its soft magnetic characteristics, the film is annealed without a magnetic filed or in a magnetic field, and then slowly cooling the film [2], [6]. Thus the CoFeHfO package-compatible magnetic material can be exploited for integration of magnetic passive components into packages.

The CoFeHfO amorphous oxide films on the substrate were simulated using the High Frequency Structure Simulator program (HFSS - Ansoft Technologies). The exposure field was set to have uniform variation on the coordinate axes, variation which can be vanished on demand. The propagation field can be chosen independent of the anisotropy axes of the thin film material, which is an advantage of the method. Three different propagation directions of the testing field were simulated, in order to verify the similitude of the results and relative errors lower than 2% were obtained.

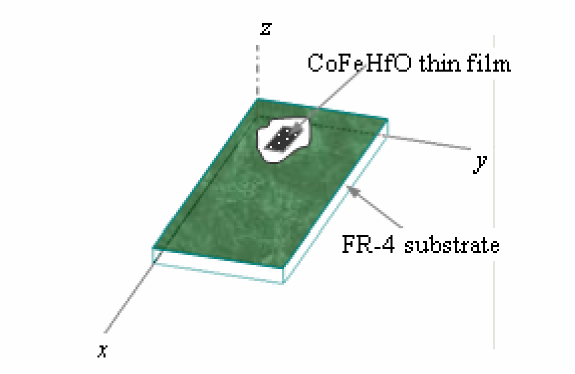

Fig. 1. A thin film of granular CoFeHfO magnetic alloy deposited on a halogen-free FR-4 substrate area.

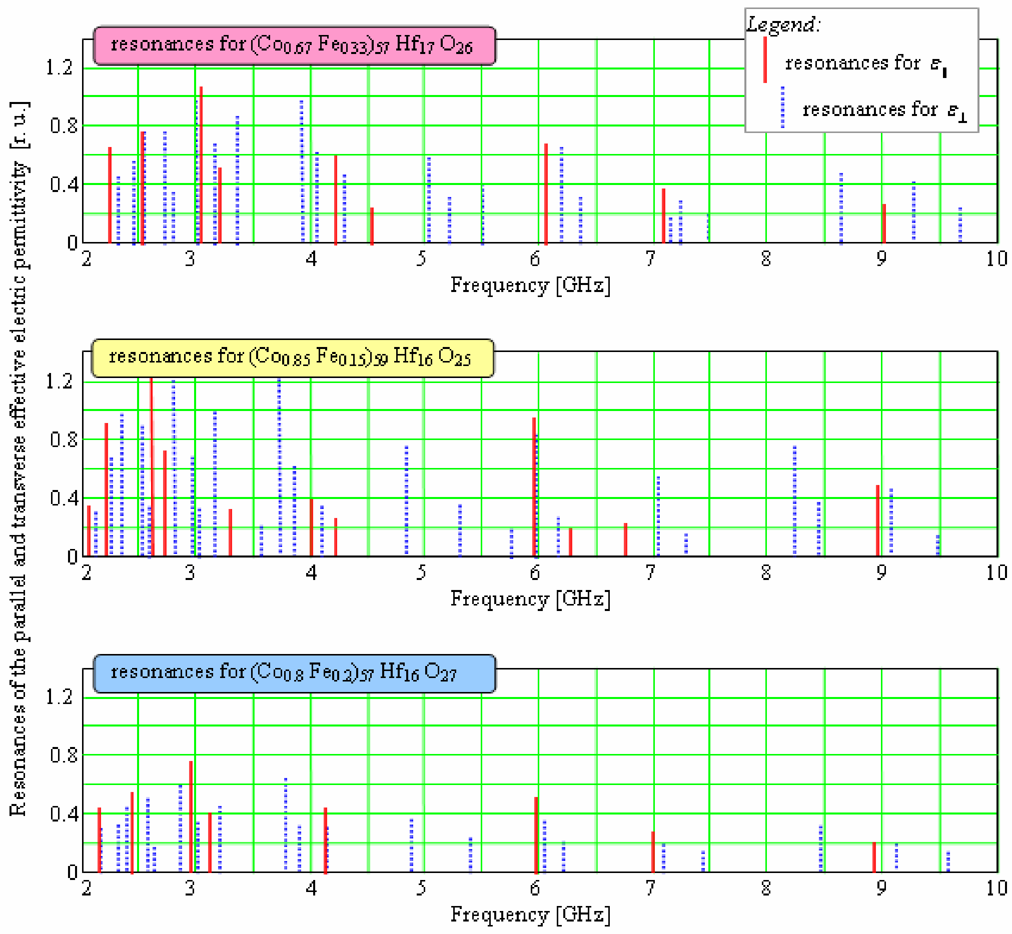

Fig. 2. Resonances of the parallel and transverse effective electric permittivity, for the 10 nm thin films of CoFeHfO magnetic alloys, deposed on a halogen-free FR-4 substrate.

Three compositions of the CoFeHfO magnetic alloy, used in practice, were analyzed by us, as follows:

- $(Co_{0.67}\ Fe_{0.33})_{57}\ Hf_{17}\ O_{26}$
- $(Co_{0.85}\ Fe_{0.15})_{59}\ Hf_{16}\ O_{25}$
- $(Co_{0.8}\ Fe_{0.2})_{57}\ Hf_{16}\ O_{27}$.

Alloys present similar but not identical electric and magnetic properties and different content of each structural element helps us to link their behavior to their composition.

The first results presented by us refer to the resonant electric behavior of the samples. Resonances of the effective electric permittivity were represented in relative units (r. u.) on the frequency scale, in the frequency domain of 2.45 - 9.8 GHz, where the accuracy of the method is the best [10], [12]. (The HFSS relates the determined resonances magnitude to the pure substrate resonances - in our case, the halogen-free FR-4). The simulation method enables us to represent separately resonances of the parallel, respective transverse electric permittivity for the anisotropic material (Fig. 2).

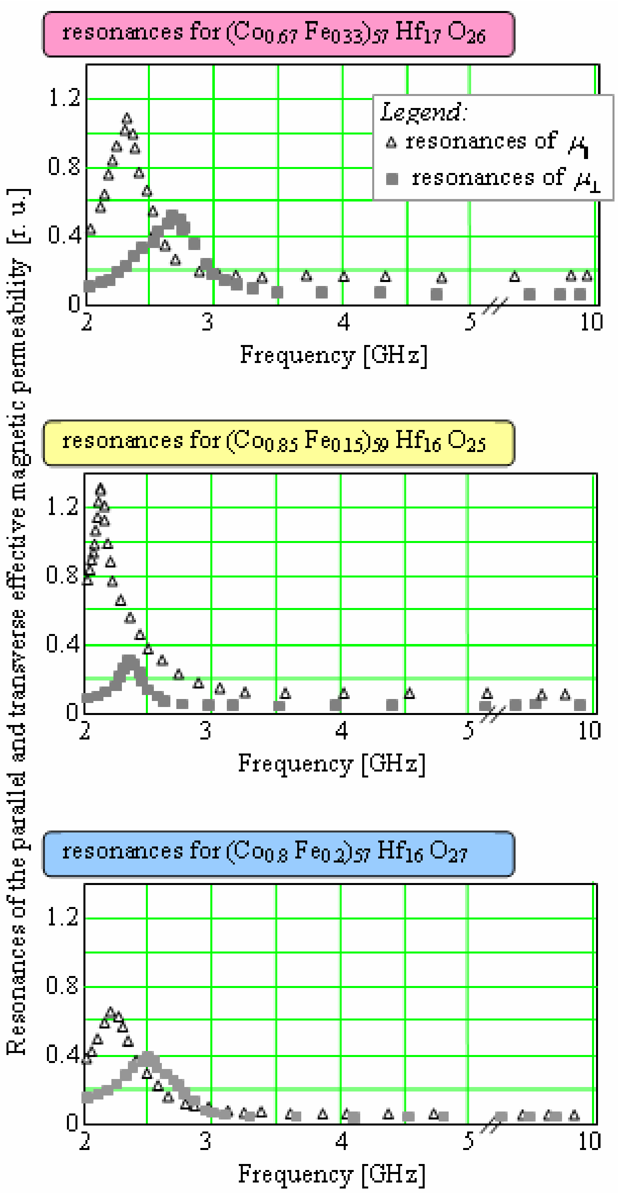

Fig. 3. Resonances of the parallel and transverse effective magnetic permeability, for the 10 nm thin films of CoFeHfO, deposed on a halogen-free FR-4 substrate.

For each material composition, an increased number of resonances appears for the transverse permittivity and their relative magnitude is lower, in comparison with the parallel permittivity resonances. These facts are due to the grains nature but not only. Simulation for material layers with different thicknesses indicates us that this is also a thin film effect, which intensifies when the layer thickness is comparable with grain size.

At the same time, the higher the frequency, the lower the resonance number (resonances rarefy on frequency scale). This appears to be due to the Hf presence in the alloy composition and to the weaker response of material at higher frequency fields.

Approximately the same number of parallel permittivity resonances was found for each material composition, resonances placed on approximately the same positions on the frequency scale. Comparing the three successive graphs, one observes that the second alloy composition presents the greatest number of resonances, which differ the most in magnitude from one to another. This is due to the greatest cobalt (Co) content in alloy. The third alloy composition presents the most flat resonances, with the lowest magnitude difference between them. This is due to the increased oxygen (O) content in the alloy. The sets of transverse permittivity resonances for each alloy composition present the same characteristics.

The magnetic effective permeability of the granular CoFeHfO nano-metric thin films was also investigated and the frequency resonant character of the permeability curves was also illustrated. The results obtained for the three considered material compositions are presented in Fig. 3. Due to the magnetic anisotropy of the CoFeHfO alloys, frequency curves for parallel, respectively transverse permeability evolves separately. Parallel respective transverse permeability are defined after the $\boldsymbol{H}$ field propagation direction (similar with the parallel and transverse permittivity) and can be normalized in respect to the permeability values after the easy magnetic axis, respectively hard magnetic axis of the magnetic alloy. Concerning the absolute value of the effective permeability of a CoFeHfO thin film, this was higher than 1500 and remained constant up to 800 MHz at least [5], depending on the alloy composition.

Graphs point out that the electric and magnetic material properties are interdependent (effective permittivity present the most resonances around the frequency of 2 GHz [7], where the effective magnetic permeability presents its maximum). Comparing Fig. 2 and Fig. 3 for the three material compositions, one observes that their groups of effective permeability resonances respect the same ordering on frequency scale like in the electric permittivity case: first, at lower frequencies, we find the resonances for $(Co_{0.85}\ Fe_{0.15})_{59}\ Hf_{16}\ O_{25}$ (the second alloy), then the resonances for $(Co_{0.8}\ Fe_{0.2})_{57}\ Hf_{16}\ O_{27}$ (the third alloy), then the resonances for $(Co_{0.67}\ Fe_{0.33})_{57}\ Hf_{17}\ O_{26}$ (the first alloy). The relative magnitudes of resonances present the same ordering (the $(Co_{0.85}\ Fe_{0.15})_{59}\ Hf_{16}\ O_{25}$ resonances are the highest). The magnitudes of the effective electric permittivity resonances, for the three analyzed material compositions have the same ordering. This ordering is required by the elements responsible for magnetism (Co, Fe and Ni) and in particular by Co, whose role is to increase the uniaxial anisotropy of the nanograins. Our magnetic alloy is a multi-phase material, as we have presented before, and the same Co element can be found in the amorphous phase (containing Co and Fe). Consequently, Co is also involved in the material electric polarization process, being included in the dipoles induced by the external field.

4. Discussion

The resonant electro-magnetic behavior of the CoFeHfO nano-metric thin films was illustrated in this paper, by material parameters determination and positioning of their resonances on frequency scale. A few particular issues have appeared in our analysis and will be pointed out as follows..

The CoFeHfO material analyzed by us is a soft magnetic alloy with high magnetic anisotropy. The material exhibits high magnetic permeability in a high frequency band and a low loss. Decrease in magnetic permeability at higher frequency (GHz range) could occur [7]. This is a loss due to the occurrence of an eddy current, drastically limited when the layer thickness decreases (~nm order). The alloy thin film present high resistivity (~ hundred of mΩ·cm order) and thus exhibits a low eddy current loss within a high frequency region. Consequently, the permeability maintains high (it does not practically decrease) in the GHz band, exceeding the resonant frequency.

Material layer thickness has a strong effect on all the high-frequency (GHz band) characteristics. Resistivity increases with decreases in the film thickness. The coercivity (H_c) and anisotropy field (H_k) of the thin films increases largely with decrease of the film thickness [5]. These facts have to be considered at HF material characterization for the simulation machine.

We have also to consider that the granular CoFeHfO presents a multi-phase structure and the material parameters resonances have to be directly linked by its internal order. These resonances are physical and geometrical resonances, whose position on the frequency scale depends on the nature, geometry and relative position of the material components.

5. Conclusions

Electro-magnetic properties for the nano-metric thin films of granular CoFeHfO magnetic alloy were analyzed in this paper, for the frequency domain of 2.45 - 9.8 GHz. New technologies recommend this material like an efficient package-compatible magnetic material, with multiple applications in practice. A simulations strategy, based on physical considerations, allowed us to obtain the HF resonant evolution with frequency of the material parameters like the effective electric permittivity and effective magnetic permeability. Material samples with three different alloy compositions were simulated on a halogen-free FR-4 board and the results are available for optimization of the material exploitation.

The effective permittivity analysis relieves that:

- for each material composition, the transverse permittivity presents more resonances which have lower magnitude than in the parallel permittivity case; this is due to a thin film effect that intensifies when the layer thickness is comparable with the grain size, and also dues to grains nature;
- the higher the frequency, the lower the resonance number, due to the weaker response of material at higher frequency fields and also to the Hf presence in the alloys;
- approximately the same number of parallel/transverse permittivity resonances was found for each material composition; resonances number and magnitude differences depend on alloy composition (Co and O content);

The effective permeability analysis points out the followings:

- electric and magnetic material properties are interdependent; the most of electric permittivity resonances occur around the same frequency where the magnetic permeability presents its resonance;
- for each material composition, the group of effective permeability resonances present the same ordering on the frequency scale like in the electric permittivity case; the relative magnitudes of the resonances also have a similar ordering; this ordering is required by the elements responsible for magnetism and in particular by Co, also involved in the material electric polarization process.

References

[1] B. P. Scaife, Principles of Dielectrics, Clarendon Press, New York, Oxford Univ. Press, 78 (1998).

[2] C.C.H. Lo, J.E. Snyder, J. Leib, D. Wang, Z. Qian, J. M. Daughton, D.C. Jiles, IEEE Trans. on Magnetics **37**, 2337 (2001).

[3] M. Eßeling, Y. Luo, K. Samwer, Europhysics Letters **68**, 100 (2004).

[4] Y. Luo, M. Eßeling, K. Zhang, G. Güntherodt, K. Samwer, Europhysics Letters **73**, 415 (2006).

[5] L.V. Tho, K.E. Lee, S.H. Kim, C.G. Kim, C.O. Kim, Materials Science Forum **558-559**, 975 (2007).

[6] L. Li, S. X. Wang, K.-P. Hwang, Y. Min, M. Mao, T. Schneider, R. Bubber, J. Appl. Phys. **99**, 08M301 (2006).

[7] D. Wang, Z. Qian, J. M. Daughton, C. Nordman, M. Tondra, D. Reed, D. Brownell, J. Appl. Phys. **89**, 675401).

[8] Y. Luo, M. Esseling, A. Käufler, K. Samwer, T. Dimopoulos, G. Gieres, M. Vieth, M. Rührig, J. Wecker, C. Rudolf, T. Niermann, M. Seibt, Physical Revue B **72**, 014426 (2005).

[9] J. Huijbregtse, F. Roozeboom, J. Sietsma, J. Donkers, T. Kuiper, E. van de Riet, J. Appl. Phys. **83**, 1579 (1998).

[10] D. Ionescu, D. Dorohoi, Scientific Annales of the “Al. I. Cuza” Univ. of Iasi **2**, 255 (2002).

[11] M. N Afsar, J. R. Birch, R. N. Clarke, Proc. of the IEEE **74**, 183 (1986).

[12] D. Ionescu, B. Ciobanu, I. Radinschi, J. Optoelectron. Adv. Mater. **9**, 2608 (2007).

[13] S. E. Russek, P. Kabos, T. Silva, F. B. Mancoff, D. Wang, Z. Qian, J. M. Daughton, IEEE Trans. on Magnetics **37**, 1756 (2001).

[14] L. Li, D.W. Lee, S.X. Wang, K.-P. Hwang, Y. Min, M. Mao, T. Schneider, R. Bubber, IEEE Trans. on Magnetics **43**, 2373 (2007).

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