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# Magnetic properties of amorphous $Co_{0.74}Si_{0.26}$/Si multilayers with different number of periods

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Two sets of $[Co_{0.74}Si_{0.26}(5\ nm)/Si(s)]_n$ amorphous films were prepared by magnetron sputtering: one in the form of multilayers with the Si spacer thickness $s$ fixed at 3 nm, and the number of periods $n$ varying from 1 to 10; and another one with only two periods and $s$ varying from 3 to 24 nm (trilayers). In both series, the $Co_{0.74}Si_{0.26}$ layer thickness $t$ was fixed at 5 nm. All the samples except the one with $s$ = 24 nm demonstrate antiferromagnetic coupling. Their magnetic properties at room temperature were probed by magnetooptical transverse Kerr effect (MOTKE) and ferromagnetic resonance (FMR). The relative increase of the saturation magnetization $M_s$ (for trilayers with respect to the one with $s$ = 24 nm; for multilayers with respect to the single layer one) obtained from the FMR measurements was compared with the exchange coupling strength $H_J^{AF}$ obtained from the MOTKE studies. $H_J^{AF}$ and $M_s$ dependencies vs $n$ and $s$ were found to be very similar to each other. Possible mechanisms of this similarity are discussed.

PACS: 75.70.Cn Magnetic properties of interfaces;
75.50.Kj Amorphous and quasicrystalline magnetic materials;
**76.50.+g** Ferromagnetic, antiferromagnetic, and ferrimagnetic resonances; spin-wave resonance;
71.70.Gm Exchange interactions.

Keywords: magnetic multilayers, amorphous films, ferromagnetic resonance, antiferromagnetic coupling.

## 1. Introduction

The interlayer exchange coupling (IEC) through silicon spacer was found in Fe/Si multilayers in 1992 [1]. Since that time different ferromagnetic metal (FM)/Si multilayers were studied both experimentally and theoretically. A significant part of the results concern the case of Fe/Si multilayers, where the coupling shows both oscillatory [1] and nonoscillatory [2] behaviors and is strongly influenced by the interface structure (namely the formation of iron silicide due to diffusion) [3,4]. Even more contradictory results were obtained on Co/Si structures, where ferromagnetic, superparamagnetic, or oscillatory ferromagnetic–antiferromagnetic behaviors of the coupling were observed [5–8]. Despite several mechanisms based on tunneling, Ruderman–Kittel–Kasuya–Yosida (RKKY)-like exchange, interface bands, and spin fluctuations have been suggested to be the origin of the coupling, still there is not much of theoretical understanding for these systems.

Recently the existence of antiferromagnetic (AF) coupling in multilayers based on amorphous $Co_xSi_{1-x}$ alloys has been found [9–11]. One of the characteristics of such magnetic amorphous compounds, in addition to their well known soft magnetic behavior, is that several properties, like the saturation magnetization and uniaxial anisotropy, can be tuned by fine adjustment of the alloy composition. In the case of $Co_xSi_{1-x}$/Si multilayers, the saturation magnetization and the Curie temperature are reduced when the

Si content is increased [10], so that the strength of the usual magnetostatic coupling contributions present in any magnetic multilayer system can be tailored. Moreover, the soft magnetic behavior of these alloys allows the detection of very weak AF couplings, that could not be observed in samples based on pure Co magnetic layers. It is known from previous studies that the $Co_xSi_{1-x}$ films are amorphous for Co concentrations smaller than $x = 0.76$ and have low coercive fields (below 1 Oe for 5 nm thick films) [12]. Thus, $Co_{0.74}Si_{0.26}$/Si multilayers appear to be a good choice of the system to study the weak AF coupling since they have a relatively soft magnetic behavior to enable the detection of the appearance of a plateau on the $M(H)$ hysteresis loop around $H = 0$, a clear footprint of the AF coupling. It is also important that the AF coupling strength can be easily extracted from the $M(H)$ loop.

Ferromagnetic resonance (FMR) has an established reputation to be a powerful tool to investigate the magnetic parameters of thin films, multilayers, and patterned structures, being particularly useful to determine the contributions from different magnetic anisotropy fields. Its effectiveness has also been demonstrated in studying the exchange coupling in ferromagnetic metal (FM)/nonferromagnetic metal (NM)/FM trilayers [13]. Here the evolution of the FMR resonance field in $Co_{0.74}Si_{0.26}$/Si multilayers was studied as a function of the AF interlayer exchange strength that was controlled either by varying the Si layer thickness or the number of periods.

## 2. Experimental details

$Co_{0.74}Si_{0.26}$/Si multilayers were grown on Si substrates by dc magnetron sputtering from high purity independent Co and Si targets. The sputtering pressure was $1.0 \cdot 10^{-3}$ mbar (with a base pressure of $\sim 10^{-9}$ mbar), and the Co target was placed at normal incidence with respect to the substrate whereas Si atoms arrive at oblique incidence (~30° with respect to the substrate normal). Two series of $[Co_{0.74}Si_{0.26}(t)/Si(s)]_n$ multilayers were prepared: one with the Si spacer thickness $s$ fixed at 3 nm, and the number of periods $n$ varying from 1 to 10; and another one with only two periods and $s$ varying from 3 to 24 nm. In both series the $Co_{0.74}Si_{0.26}$ layer thickness $t$ was fixed at 5 nm. In all the cases a 3 nm thick Si buffer layer was grown on top of the native oxide of the substrate before growing the corresponding multilayer. A protective capping layer of Si of the same thickness was always deposited on top of the samples in order to prevent oxidation.

A magnetooptical transverse Kerr effect (MOTKE) system was used to study the hysteresis loops of the samples at room temperature. The MOTKE signal, $\delta K$, is defined as $\delta K = (R^+ - R^-)/R$, where $R^+$ is the reflectivity for positive applied magnetic field, $R^-$ is that for negative field, and $R$ is the value for an idealized nonmagnetized sample, in practice taken as the average of $R^+$ and $R^-$. For a thin film, $\delta K$ is proportional to the magnetooptic constant $Q$ which, in a first approximation, is linear in the saturation magnetization $M_S$.

$Co_{0.74}Si_{0.26}(t)$/Si multilayers were probed by continuous wave ferromagnetic resonance. FMR field at room temperature was measured using a standard $X$-band electron paramagnetic resonance spectrometer Bruker ESP 300 (~9.8 GHz) for the full range of angles $\theta$ between the external field direction and the normal to the film plane (0°–90°). Additionally, in-plane angular dependences (azimuthal angle $\varphi$ variation from 0° to 360° for $\theta = 90°$) of the resonance field $H_r$ were measured to determine the in-plane uniaxial anisotropy, expected in this system from previous studies [9,10].

## 3. Results and discussion

Magnetooptical studies reveal that the single layer of $Co_{0.74}Si_{0.26}$/Si has a coercive field of 0.5 Oe, and has, as all the other multilayered samples involved in the experiment, well defined uniaxial anisotropy with an anisotropy field of approximately 20 Oe. The magnetic behavior of the series of $Co_{0.74}Si_{0.26}$ (5 nm)/Si($s$ nm)/$Co_{0.74}Si_{0.26}$ (5 nm) trilayers is demonstrated in Fig. 1 and shows the clear presence of an AF coupling for Si spacer thicknesses up to 12 nm. All the samples except the one with $s = 24$ nm have an almost null remanence, presenting a clear plateau around $H = 0$. A step in the loop appears for magnetization values close to zero, which is related to the AF state of the trilayer. It is easy to notice that the coupling between the layers is monotonically decreasing with the increase of $s$ and completely disappears above 20 nm. The coupling field strength $H_J^{AF}$ for the trilayer samples was extracted from the simple formula (proposed in Ref. 10)

$$H_J^{AF} = \frac{H_3 - H_1}{2}, \quad (1)$$

where $H_1$ is the field where the reversal process starts the formation of an AF coupled state when the external field is continuously decreasing from the maximum positive value,

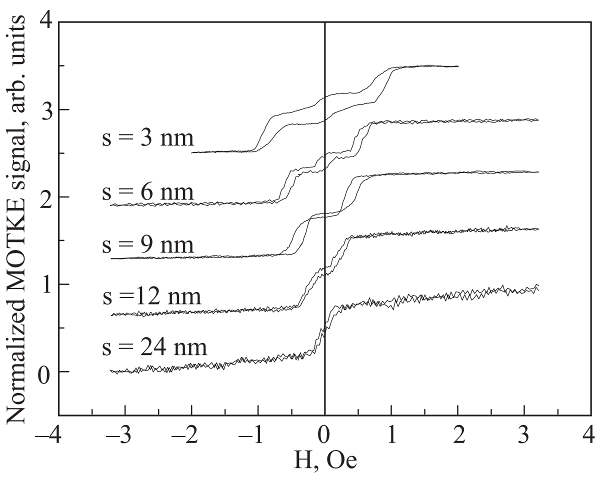


*Fig. 1.* Normalized MOTKE hysteresis loops for $[Co_{0.74}Si_{0.26}(5\ nm)/Si(s)]_2$ with different thicknesses of the Si layer $s$. The curves have been vertically shifted for clarity.

and $H_3$ is the field where the AF state suddenly breaks leading to a fast change in magnetization up to values close to the negative saturation.

For the case of the strongest AF coupling ($s$ = 3 nm), the magnetic evolution of the MOTKE hysteresis loop with increasing number of periods of $[Co_{0.74}Si_{0.26}$ (5 nm)/Si(3 nm)$]_n$ is presented in Fig. 2. As the number of periods is increased, other steps appear, although their field width decreases continuously, as well as the whole process of the magnetization reversal between both saturated states keeps smoothing, as it is shown in Fig. 2 for the case of $n$ = 6 (note that the saturation field is much smaller than the anisotropy field of 20 Oe obtained from the completely closed hard axis hysteresis loops). None of the samples have zero MOTKE signal at zero field, but most are very close to it. For our case of only even number of periods the signal takes no zero values even for a zero net magnetization mainly due to interference effects. One can observe that with the increase of the number of periods the strength of the interlayer interactions is increasing, saturating at $n$ = 6–8. For multilayered samples the coupling field strength $H_J^{AF}$ was extracted from the formula similar to Eq. (1):

$$H_J^{AF} = \frac{H_4 - H_1}{2}, \qquad (2)$$

where $H_4$ is the field at which the magnetization reaches negative saturation. Whereas for the trilayers the values of $H_3$ and $H_4$ are almost identical and it is easy to determine $H_3$, for multilayers only $H_4$ can be clearly determined.

The FMR data were fitted using the well-known Kittel equation with two parameters, the $g$-factor and the effective anisotropy field, $H_{\text{eff}} = 4\pi M_s - H_\perp$, where $M_s$ is the saturation magnetization and $H_\perp$ is the sum of all possible perpendicular anisotropies (see Ref. 14 for more details). In the case of ferromagnetic/nonmagnetic multilayers, the expected contributions to $H_\perp$ are as follows: the surface-related ($H_s$), magnetocrystalline ($H_k$) and magnetoelastic ($H_\sigma$) terms, and the one due to the indirect exchange between the neighboring layers ($H_{\text{ex}}$).

In-plane FMR angular dependence measurements confirmed the presence of a weak in-plane uniaxial anisotropy in all the samples (detected as well by MOTKE studies), with approximately the same anisotropy field value $H_{\text{in-plane}} \approx 20$ Oe. We would like to remind that except very special cases of strong in-plane uniaxial anisotropy (i.e., when $H_{\text{in-plane}}$ is comparable with resonance field, see, for example, Ref. 15) $H_{\text{in-plane}}$ can be easily extracted from the Eq. (6) of Ref. 14. For the single period $Co_{0.74}Si_{0.26}$ film, the best fit to the out-of-plane $H_r(\theta)$ dependence was obtained with the effective anisotropy field $H_{\text{eff}} \approx 0.9$ kOe that corresponds to $M_s \approx H_{\text{eff}}/4\pi = 73$ G, in a good agreement with the previous studies [9,10]. When going from the single layer film to the trilayer one ($n$ = 2), with a Si interlayer thickness $s$ = 3 nm, the effective anisotropy field increases by 100 Oe. With the increase of the Si interlayer thickness for $n$ = 2, the effective anisotropy is decreasing down to the value of the single layer film $H_{\text{eff}} \approx 0.9$ kOe for $s$ = 24 nm (see Fig. 3). For the second series ($s$ is fixed at 3 nm, $n$ varies from 2 to 10), the effective anisotropy gradually increases up to 1050 Oe, reaching the saturation at $n$ = 8 (see Fig. 4).

The obtained relative changes of the effective anisotropy (for trilayers with respect to the one with $s$ = 24 nm; for multilayers with respect to the single layer one) were compared with the exchange coupling strength $H_J^{AF}$ values (see Fig. 5 and 6, respectively). One can observe that $H_J^{AF}$ and $H_{\text{eff}}$ dependencies in both cases are very similar to each other. This fact allows us to suggest that the exchange coupling strength, despite being very weak, significantly affects the effective anisotropy of the system: for the $[Co_{0.74}Si_{0.26}$(5 nm)/Si(3 nm)$]_{10}$ multilayer the effective anisotropy is ~17% bigger than for the single layered film. There are several possible mechanisms that could change the effective anisotropy of the system. As it was mentioned

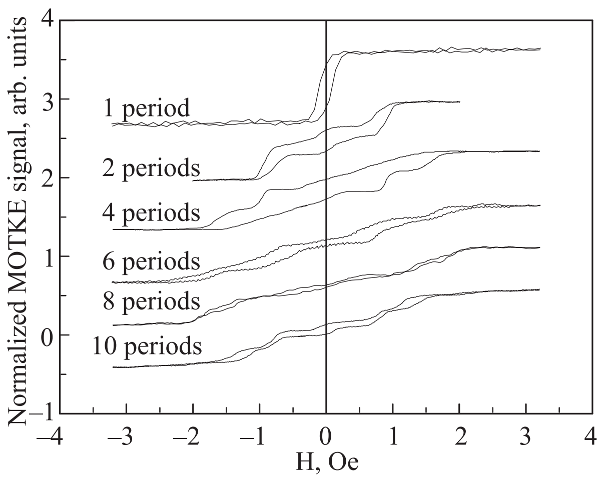


*Fig. 2.* Normalized MOTKE hysteresis loops for $[Co_{0.74}Si_{0.26}$(5 nm)/Si(3 nm)$]_n$ with different numbers of periods. The curves have been vertically shifted for clarity.

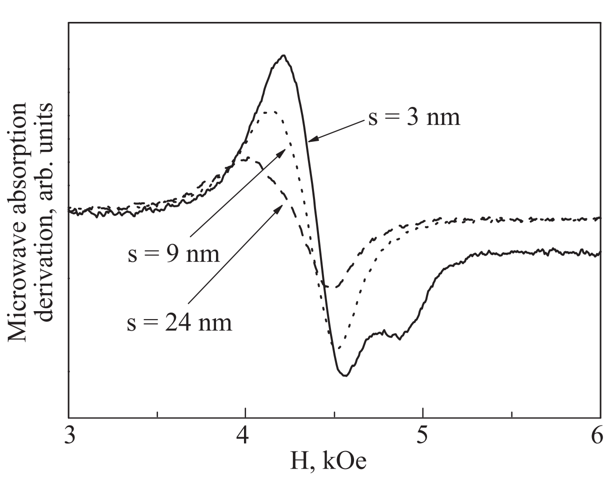


*Fig. 3.* Ferromagnetic resonance signals (first derivative of the microwave absorption) at θ = 0° for $[Co_{0.74}Si_{0.26}$(5 nm)/Si($s$)$]_2$ with different thicknesses of the Si layer $s$.

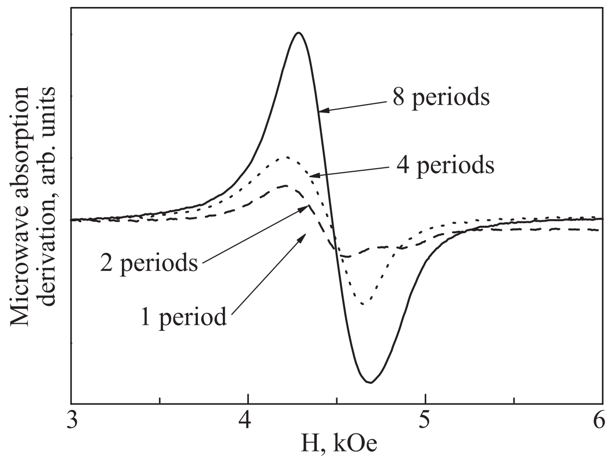


*Fig. 4*. Ferromagnetic resonance signals (first derivative of the microwave absorption) at $\theta = 0°$ for $[Co_{0.74}Si_{0.26}(5\ nm)/Si(3\ nm)]_n$ with different numbers of periods.

above, the effective anisotropy consists of the shape anisotropy (that is proportional to the saturation magnetization), the surface-related ($H_s$), magnetocrystalline ($H_k$) and magnetoelastic ($H_\sigma$) contributions, and the one due to the indirect exchange between the neighboring layers ($H_{ex}$). In case of amorphous magnetic layers, $H_k$ and $H_\sigma$ should be negligibly small as compared to $4\pi M_s$, and $H_s$ at the given thickness of ferromagnetic layer $t = 5$ nm is also very small and independent of $n$, so that all these contributions can be excluded from the further discussion. Exchange coupling $H_{ex}$ could also modify $H_{eff}$, however, direct calculations of $H_{ex}$ contribution using formulas from Ref. 13 demonstrate that even for the strongest coupling case (i.e., $Co_{0.74}Si_{0.26}(5\ nm)/Si(3\ nm)]_{10}$ sample) the additional contribution to $H_{eff}$ will be ~ 10 Oe, i.e., 17 times less than the value found from the FMR experiments. Even more, in case of an AF exchange coupling strong enough to noticeably change the FMR resonance fields, the $H_r(\theta)$ angular dependence will change in a different way than observed here: both for in-plane and perpendicular-to-plane configurations the $H_r$ values will shift to higher fields comparing to the noninteracting sample. As a result, the formal analysis using Kittel formula will demonstrate a change of the $g$ factor rather than in $H_{eff}$. Actually, the $g$ factor was almost the same (≈ 2.15) for all the samples included in this study. Thus, the influence of $H_{ex}$ on effective anisotropy appears to be negligible.

Therefore, after all other terms have been excluded, the only possible explanation left for the observed enhancement in $H_{eff}$ is the change of the saturation magnetization as a function of either the Si spacer thickness for the trilayers or the number of periods for the multilayers. This change in $M_s$ is proportional to the exchange coupling strength in the sample. Two possible factors could modify the room temperature $M_s$ in these multilayers: First, the variation of the Si spacer thickness of the trilayer may change the Si diffusion rate from the spacer to ferromagnetic layers and, as a result, the Co content and magnetization of the trilayers with different $s$ values will change gradually. However, this scheme cannot explain the gradual increase of the magnetization in the multilayered samples with increasing period numbers. Also, in the case of interdiffusion, the Co concentration will gradually change from the border to the center of the layer. This should increase the FMR linewidth of the samples with lower $H_{eff}$ (i.e., saturation magnetization). However, no significant and systematic change of the FMR linewidth was found in the samples under study. Second, it is known that for $Co_{0.74}Si_{0.26}$ the Curie point is close to the room temperature. Then if the exchange coupling can change $T_C$ even slightly, it still can lead to a noticeable change of the room temperature magnetization which could explain all the obtained results for both series.

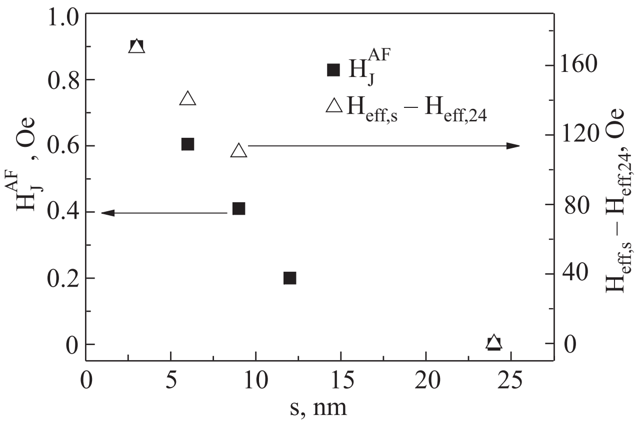


*Fig. 5.* Comparison of the exchange field strength (obtained from MOTKE measurements) and change in the effective anisotropy field with respect to the sample with $s = 24$ nm (obtained from FMR measurements) for trilayers with different Si layer thickness.

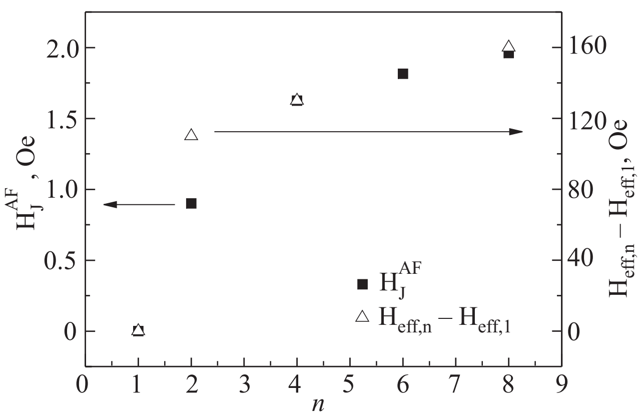


*Fig. 6.* Comparison of the exchange field strength (obtained from MOTKE measurements) and relative change in effective anisotropy field with respect to the single layer sample (obtained from FMR measurements) for multilayers with different number of periods.

## Conclusions

The exchange coupling strength $H_J^{AF}$ in $[Co_{0.74}Si_{0.26}(5\ nm)/Si(s)]_n$ was tuned either by varying the Si spacer thickness *s* for the $[Co_{0.74}Si_{0.26}(5\ nm)/Si(s)]_2$ series or by changing the number of periods *n* for the $[Co_{0.74}Si_{0.26}(5\ nm)/Si(3\ nm)]_n$ series. The relative increase of the effective anisotropy field and, the corresponding saturation magnetization (for trilayers with respect to the one with $s = 24$ nm; for multilayers with respect to the single layer one) obtained from FMR measurements was found to be proportional to $H_J^{AF}$.

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1. S. Toscano, B. Briner, H. Hopster, and M. Landolt, *J. Magn. Magn. Mater*. **114**, L6 (1992).
2. E.E. Fullerton, J.E. Mattson, S.R. Lee, C.H. Sowers, Y.Y. Huang, G. Felcher, and S.D. Bader, *J. Magn. Magn. Mater*. **117**, L301 (1992).
3. R.R. Gareev, D.E. Bürgler, M. Buchmeier, R. Schreiber, and P. Grünberg, *Appl. Phys. Lett*. **81**, 1264 (2002).
4. J.M. Pruneda, R. Robles, S. Bouarab, J. Ferrer, and A. Vega, *Phys. Rev*. **B65**, 024440 (2001).
5. P.J. Grundy, J.M. Fallon, and H.J. Blythe, *Phys. Rev*. **B62**, 9566 (2000).
6. J.M. Fallon, C.A. Faunce, and P.J. Grundy, *J. Appl. Phys*. **88**, 2400 (2000).
7. K. Inomata and Y. Saito, *J. Appl. Phys*. **81**, 5344 (1997).
8. T. Luciński, P. Wandziuk, F. Stobiecki, B. Andrzejewski, M. Kopcewicz, A. Hütten, G. Reiss, and W. Szuszkiewicz, *J. Magn. Magn. Mater*. **282**, 248 (2004).
9. C. Quirós, J.I. Martin, L. Zarate, M. Velez, and J.M. Alameda, *Phys. Rev*. **B71**, 024423 (2005).
10. L. Zárate, C. Quirós, M. Vélez, G. Rodríguez-Rodríguez, J.I. Martín, and J.M. Alameda, *Phys. Rev*. **B74**, 014414 (2006).
11. L. Zárate, C. Quirós, M. Vélez, J.I. Martín, and J.M. Alameda, *J. Non-Cryst. Solids* **353**, 959 (2007).
12. M. Vélez, C. Mény, S.M. Valvidares, J. Díaz, R. Morales, L.M. Alvarez-Prado, and J.M. Alameda, *Eur. Phys. J*. **B41**, 517 (2004).
13. J. Lindner and K. Baberschke, *J. Phys: Condens. Matter* **15**, 193 (2003).
14. G.N. Kakazei, P.E. Wigen, K.Yu. Guslienko, R. Chantrell, N.A. Lesnik, V. Metlushko, H. Shima, K. Fukamichi, Y. Otani, and V. Novosad, *J. Appl. Phys*. **93**, 8418 (2003).
15. G.N. Kakazei, A.F. Kravetz, N.A. Lesnik, M.M. Pereira de Azevedo, Yu.G. Pogorelov, G.V. Bondarkova, V.I. Silantiev, and J.B. Sousa, *J. Magn. Magn. Mater*. **196–197**, 29 (1999).