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The effect of Microstructure in Exchange Decoupling of $SmCo_5$/Co bi-layers at low temperatures

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Abstract:*Here, we investigatedlow temperature magnetic properties of $SmCo_5$/Co bilayer samples on MgO(100) and glass substrates. Samples were fabricated under identical conditions with a 60 nm Cr underlayer and magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer with the maximum applied field of 7 T. Analysis of each layer by an Atomic Force Microscope (AFM) reveals that MgO(100) results in small and uniform $SmCo_5$grain formation in contrast to glass. X-ray diffraction studies show that the sample on MgO(100) has high crystallinity with $SmCo_5(11\bar{2}0)$ phase. Atroom temperature, both samples exhibit good hard magnetic properties withcoercivities of 13.2 kOe and 12.5 kOe, depositedon MgO(100) and glass, respectively.Low temperature hysteresis measurements show a developmentof an exchange decoupling phenomenon below 150 K for the sample on glass, and we propose that this is due tothe formation of large magnetic grains on glass that reduces the effective inter-grain exchange coupling between soft and hard magnetic phases.*

I. Introduction

Exchange-coupled magnets that are composites of soft and hard magnetic materials have been explored since its inception by Kneller and Hawig in 1991[1].This new magnet family has many intriguing properties such as high remanence (M_r), high energy products $(BH)_{max}$, high curie temperatures (T_C)and lower cost [1-4], which made them ideal candidates to replace existing hard magnetic materials for a wide range of applications from data storage to energy efficient appliances. In these magnets, the coercivity (H_C) and M_r are determined by the hard magnetic and the soft magnetic phases, respectively, while effective exchange couplingbetween the phases holds the key to achieving optimum magnetic properties. This exchange couplingacross the soft-hard magnet interface depends not onlyon materials and their physical dimensions but also on the grain size and distribution of each phase, which made it one of the poorly understood phenomenon in exchange-coupled systems[5].In addition, in thin films, substrate plays a key role in controlling microstructurewhich can come as epitaxial guidance, epitaxial mismatch or even de-wetting at given processing conditions, making the microstructure is unique to thesubstrate[3,6-10]. As a result, samplesdeposited on different substrates can have very different magnetization reversal paths,producing different magnetic properties even for the same material combination. As an example, Chowdhury *et. al.*[6] reported that $SmCo_5$/Co bi-layers grown on MgO(110) and Si(100) under identical processing conditions with same layer thicknessresulted energy products of 20.1 MGOe and 12.4 MGOe, respectively.

Here, we studied the reversal of $SmCo_5$/Co exchange-coupled bi-layer thin films in the 300-50 K temperature regime, deposited onMgO(100) and glass substrates under identical conditions. We choseMgO(100) and glass as substrates, as previous studies reveal that both these substrates result in high H_C $SmCo_5$ thin films[7,8]at room temperature. This study extends beyond the previous work by investigating magnetic properties of $SmCo_5$/Co bilayers at temperatures below the room temperature and explored how these properties are affected by the substrate. Room temperature magnetic measurements confirm spring-exchange behavior with high H_C for both samples, however, low temperature hysteresis measurements show an exchange decoupling like phenomenon for the sample deposited on glass. This transformation from single-step to two-step hysteresis,below a critical temperature, suggests a weakening of exchange coupling.

II. Sample Preparation

$SmCo_5$/Co exchange spring bi-layer films were fabricated ina DC and RF magnetron sputtering system (Orient 8, AJA Inc.) at high vacuum of 10^{-8}Torr on MgO(100) (sample A) and glass (sample B)substrates, which were attached to a rotating stage equipped with a heater. Thickness of each layer was fixed at 60 nm, 30 nm and 7.5 nm for the Cr seed layer, $SmCo_5$ and Co layers, respectively. The nominal thickness of $SmCo_5$ and Co was determined based on highest $H_C$at room temperature with single-step hysteresis. An alloy target with the proper composition was used to deposit $SmCo_5$, and both the seed and $SmCo_5$layers were sputter deposited at 500°C that is adequate enough to induce in-plane hard magnetic properties of $SmCo_5$ layer [11,12]. After the growth of Cr and $SmCo_5$ layers, samples were allowed to cool for 6 hours before depositing Co layer. This is an important

step to minimize inter-diffusion at the $SmCo_5$/Co interface, which can change the composition of the hard phase. A 30 nm Cr layer was deposited onto Co layer to protect magnetic layers from oxidation. To investigate grain formation of Cr and $SmCo_5$layers, a set of Cr and Cr/$SmCo_5$ samples were fabricated on both substrates under identical fabrication conditions as of sample A and B.All layers were sputter deposited at 4 mTorrin Arenvironmentand low sputtering powers were used to keep low deposition rates that promotes continues film growth.

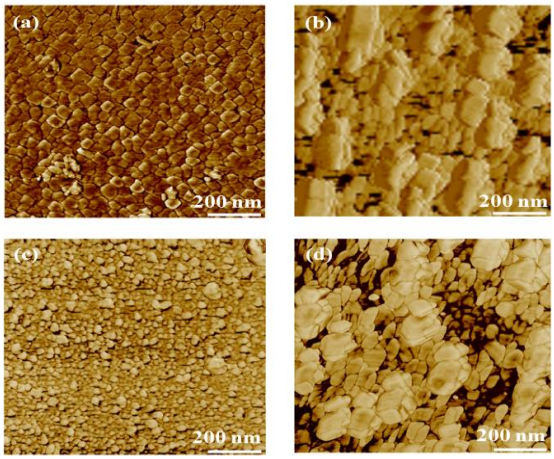

FIG. 1.AFM micrograph (phase mode) of Cr (60 nm) underlayer grown at 500 ^{0}C on (a) MgO(100) and (b) glass, and $SmCo_5$(30 nm) grown at 500 ^{0}C on (c) MgO(100) and (d) glass substrates with a 60 nm Cr underlayer.

III. Results and Discussion

The grain size and distribution of each layer was analyzed by an AFM (VeccoNanoscope IV, Bruker Inc.) and their crystallinitywasinvestigated by x-ray diffraction with Cu K_α radiation (X'Pert MRD, Panalytical Inc.). Fig. 1 shows the AFM micrographs of 60 nm Cr underlayer,and 30 nm $SmCo_5$ hard magnetic layer deposited on Cr underlayer at 500 ^{0}C on MgO(100) and glass. As fig. 1(a) and (b) depict, Cr layer deposited on MgO(100) has a nice texturewith square shape grains, as opposed to large inhomogeneous Cr grains on glass. This highly textured Cr grain formation might be a result of epitaxial guidance by MgO(100), as reported in previous studies [3, 4].The average sizes of Cr grains are26 (± 8) nmand 88 (± 54) nm on MgO(100) and glass, respectively.As shown in fig. 1(c) and (d), $SmCo_5$grain formation is unique to the substrate, as Cr/MgO(100) results smaller $SmCo_5$ grains with high degree of uniformity while Cr/glass produces large and inhomogeneous $SmCo_5$ grains.The average grain sizes are 19 (±7) nm and 34 (±32) nm on Cr/MgO(100) and Cr/glass, respectively. These measurements confirm MgO(100) drives small and uniform grain formation in contrast to grains on glass. Previous studies show that small grains andhigh volume of grain boundaries are crucial for effective interfacial exchange coupling [13, 14].

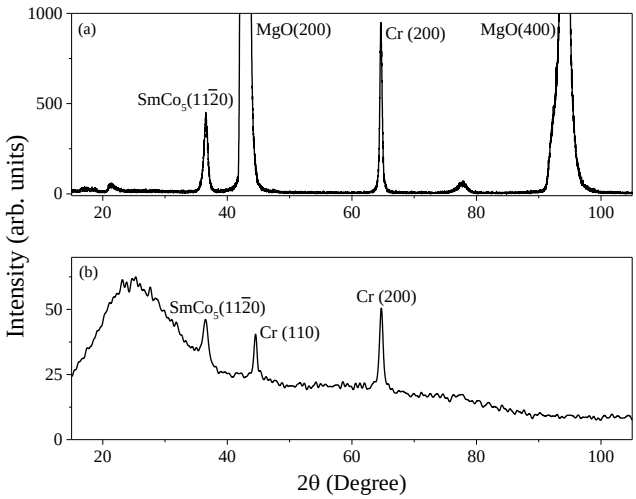

FIG. 2. XRD patterns of Co/$SmCo_5$/Cr films sputtered onto (a) MgO(100), and (b) glass. The growth temperature was set to 500 ^{0}C for $SmCo_5$ and Cr layers.

Fig. 2 showsx-ray diffraction patterns of sample A(a) and B (b), respectively. For sample A, strong diffraction peaks corresponds to MgO(200), Cr(200) and $SmCo_5(11\bar{2}0)$ planes can be observed. The absence of other Cr phases shows that MgO(100) promotesepitaxial growth of Cr(200) that guides the growth of highly textured $SmCo_5(11\bar{2}0)$ phase[3,4].However, the diffraction pattern of sample B shows the presence of bothCr(110) and Cr(200) phases, yet the peak intensity of Cr(110) is roughly one half of the Cr(200) peak. Since Cr(110) is the dominating crystalline phase for isotropic Cr samples grown at room temperature [8, 15], this strong signal for Cr(200)confirms that the Cr(200) phase is mainly driven by high temperature annealing.The formation of $SmCo_5(11\bar{2}0)$ crystalsin sample B, guided by Cr(200) phase, shows that in-plane hard magnetic properties are not unrealistic even on amorphous substrates such asglass with a matching buffer layer and proper growth conditions. However,the low intensity of $SmCo_5(11\bar{2}0)$ peak assures that most of the $SmCo_5$ are amorphous. In-plane magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer (MPMS-7T, Quantum Design Inc.) with a maximum field of ±7 T.Fig. 3 shows normalized hysteresis of both sample A and B measured at 300 K.The $H_C$of sample A and B are 13.2 kOe and 12.5 kOe, respectively, suggesting that $SmCo_5(11\bar{2}0)$ may be responsible for high H_C in both samples[16-19].Although both samples show competitive H_C values, the sample A has higher$(BH)_{max}$ of 14.5 MGOe in contrast to 5.3 MGOe of sample B.The low $(BH)_{max}$ of sample B can bedue to low in-plane moment, caused by therandom orientation of $SmCo_5$ grains. Thisis even reflected in its x-ray diffraction pattern with a weak signal for $SmCo_5(11\bar{2}0)$. As a result, highH_C does not necessarily guarantee a high $(BH)_{max}$ as random orientation of crystals significantly lowers the effective magnetizationand hence the maximum energy product, which is given by[1],

$$(BH)_{\max} = \frac{1}{4}\mu_o M_S^2 \quad \text{..... (1)}$$

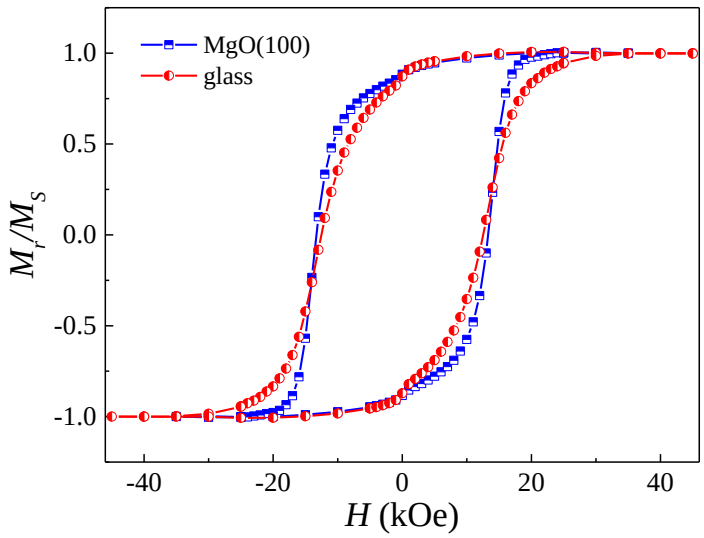

FIG. 3. Normalized room temperature hysteresis curves of$SmCo_5$/Cobilayers on MgO(100) and glass substrates grown at 500° C.The sample on MgO(100) shows higher degree of squareness.

The temperature dependent magnetic properties of these two samples were measured from 300 - 50 K and corresponding hysteresis curvesfor sample A and B are shown in fig. 4(a) and 4(b), respectively. Table 1 depicts the data extracted from fig. 4(a) and 4(b) at various temperatures from 300 K-50 K. For the sample A, $H_C$increases linearlyfrom 13.2 kOe at 300 K to 23.3kOe at 50 K while $M_S$largely remains at 2.42x10^{-4} emu for all temperatures, except for a slight drop at 100 K and 50 K (Table 1). The reduce remanence M_r/M_S fluctuates between 0.88-0.90 with no clear trend with the temperature.It is remarkable to see that at every temperature, the single step hysteresis is preserved. This implies that theexchange coupling between soft and hard phases is preserved at low temperatures. The increase in $H_C$can be ascribed to increase in effective magnetocrystalline anisotropy and higher degree of pinning at lower temperatures. However, it can be observed that there is a slight decrease in the squareness of hysteresis loops with lowering the temperature.

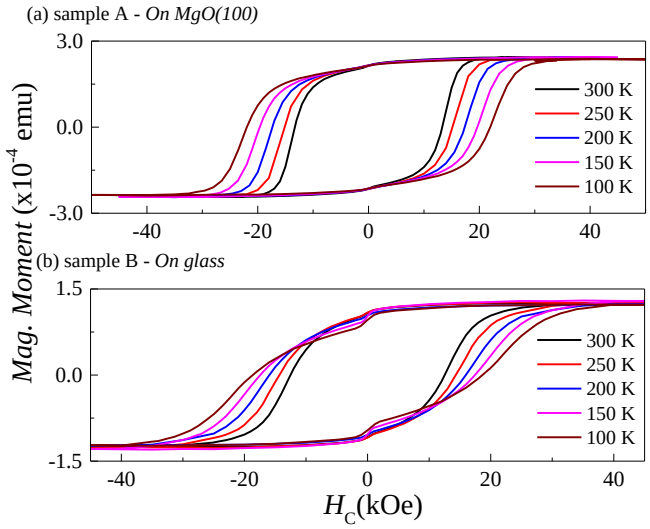

FIG. 4.Low temperature hysteresis curves measured at 300 K (black), 250 K (red), 200 K (blue), 150 K (magenta), 100 K (brown) for the $SmCo_5$/Co sample on (a) MgO(100) and (b) glass. Formation of a 'shoulder' can be seen in hysteresis curves in graph (b) for 150 K and 100 K.Note: 50 K hysteresis is not shown for clarity.

For the sample B, the $H_C$increases from 12.5 kOe at 300 K to 21.8kOe at 50 K whilethe saturation moment shows a random variation with the temperature as shown in Table 1.The reduced remanence M_r/M_S remainsat 0.87-0.88 for 300-200 K temperature regime, however a considerable drop from0.88 to 0.80 can be seen when reducing the temperature from 200 K to 150 K. This drop in M_r/M_S coincides with the formation of a 'shoulder' in 150 K, 100 K and 50 K (50 K measurement is not shown in fig. 4) hysteresis curves. This transformation from single-stepto two-step hysteresisindicates a decoupling of soft and hard phases below a critical temperature. This phenomenon has beenpreviously observed and accountedfor exchange decoupling that takes place when lowering the temperature[20, 21] as follows. Based on first principle calculations, effective exchange coupling between soft and hard phases and the single-step reversal require the soft phase to be confinedto thesize of domain wall width of the hard phase (δ_K)[1-3]. However, δ_K is governedbythe effective anisotropy K, as $\delta_K \propto 1/\sqrt{K}$, which increases with decreasing the temperature. This makesδ_K drops when decreasing the temperature, mandating a smaller soft region to keep the exchange coupling intact at lower temperatures. Since the physical size of the soft region remains unchanged, decreasing the temperaturemakesthese two phases partially or fully decoupled,resultinga two-step hysteresis.However, here, we only see such decoupling for sample B despite both samples have identical soft and hard layer thickness and fabricated under identical conditions. Topographic analysis of these samples (Fig. 1) shows that sample A has small and uniform grains in contrast to sample B, which has large grains and inhomogeneous size distribution. Thissuggests that exchange decoupling observed for sample Bmay have been caused by its microstructure.Theidea of grain-controlled magnetic properties of thin films can be supported by a number of recent studies that propose smaller grains and uniform distribution favor large inter-grain exchange couplings that enhance the remanence[22-24]. This is in-line with higher remanence and higher $(BH)_{max}$ of sample A in contrast to that of sample B. Another key observation in hysteresis of sample B is that there is a cross-over betweenlow temperatureand hightemperature curves (Fig. 4 (b)). This indicates achange of the reversal mechanism due to the exchange decoupling between hard-soft phases that results a weakening of hard magnetic properties.

Table 1. Coercivity (H_C), saturation moment and reduced remanence (M_r/M_S) for sample A and sample B for temperatures between 300 K – 50 K (extracted from fig. 4)

Temperature (K)	Sample A			Sample B		
	H_C (kOe)	*Sat. Moment* (x10^{-4} emu)	M_r/M_S	H_C(kOe)	*Sat. Moment* (x10^{-4} emu)	M_r/M_S
300	13.2	2.42	0.88	12.5	1.24	0.87
250	15.1	2.41	0.90	14.3	1.26	0.87
200	17.3	2.42	0.89	15.9	1.23	0.88
150	19.6	2.42	0.88	17.4	1.28	0.80
100	22.0	2.38	0.90	18.6	1.23	0.80
50	23.3	2.39	0.89	21.8	1.29	0.79

Fig. 5 shows the variation of the reduced remanence (M_r/M_S) and coercivity (H_C) with the temperature. As shown in Fig. 5(a), $H_C$increases almost linearly with the temperature with slightly different slopes of -0.042 and -0.035 with extrapolated coercivities of25.76 kOe and 22.84 kOe at 0 Kfor sample A and B, respectively. This steep increase in $H_C$when lowering the temperature for sample Acan be associated with large concentration of domain wall boundaries resulted by smaller grains, compared to those of sample B. Further, fig 5(b) shows thatM_r/M_Shas no significant temperature dependence for sample A, however, a considerable dropfrom 0.88 to 0.80 for sample B can be observed when the measuring temperature is reduced from 200 K to 150 K. This could be a result of weakened exchange coupling between soft and hard magnetic grainsthat essentially reduces the remanence.

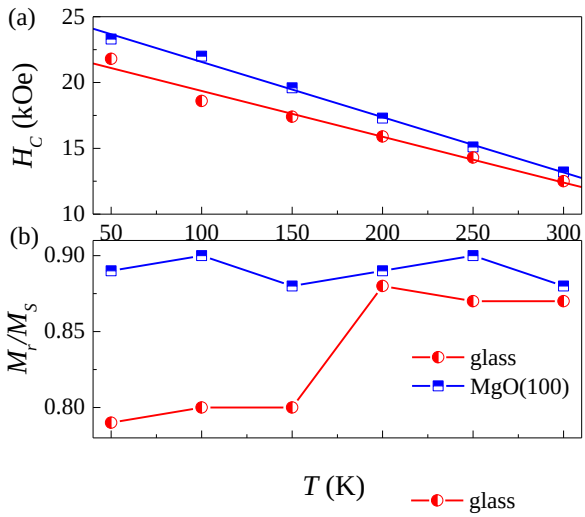

FIG. 5. Variation of (a) coercivity (H_C), and reduced remanence (M_r/M_S) with temperature for sample A (blue) and sample B (red). $H_C$was fitted to linearfunctions with corresponding slopes of -0.042 and -0.035 for sampleA and sample B, respectively.

IV. Conclusions

In this work, we investigated magnetization reversal of exchange-coupled magnetic thin films fabricated on single crystal MgO(100) and amorphous glass substrates with a 60 nm Cr seed layer. X-ray diffraction studies show that in-plane hard magnetic properties are due to the formation of twisted-crystalline $SmCo_5$ $(11\bar{2}0)$ phase, guided by the Cr (200) seed layer. AFM measurements of each magnetic layer reveals that MgO(100) induced small and uniform grains of 19 (±7) nm in contrast to larger grains on glass with a random size distribution, 34 (±32) nm. Room temperature hysteresis measurements show that both samples exhibit good hard magnetic properties with high coercivitiesbut $(BH)_{max}$ of the sample on glass (5.3 MGOe) is almost 1/3 of that of the sample on MgO(100) due to amorphous nature and random orientation of crystals that lowers the effective in-plane magnetization. Hysteresis measurements at lower temperaturesreveal an exchange decoupling like phenomenon only for the sample on glass.We believe that this decoupling is induced by the microstructure, as large magnetic grains on glass substrate reduce the effective inter-grain exchange coupling between soft and hard magnetic phases, which is critical against rising anisotropy when lowering the temperature.

Acknowledgments

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Reference

[1]. Kneller, F., Reinhard, H., *Magnetics, IEEE Transactions on*27.4 (1991): 3588-3560.
[2]. Skomski, R., Coey, J. M. D., *Physical Review B* 48.21 (1993): 15812.
[3]. Fullerton, E. E., Jiang, J. S., Bader, S. D.,*Journal of Magnetism and Magnetic Materials* 200.1 (1999): 392-404.
[4]. Fullerton, E. E., Jiang, J. S., Grimsditch, M., Sowers, C. H., Bader, S. D.,*Physical Review B*, *58*(18), (1998): 12193.
[5]. Xiao, Q. F., Zhao, T., Zhang, Z. D., Brück, E., Buschow, K. H. J., & de Boer, F. R., *Journal of magnetism and magnetic materials*, *223*(3), 215-220, (2001).
[6]. Chowdhury, P., Krishnan, M., Barshilia, H. C., Rao, D. S., Kumar, D., Shivakumara, C.,*Journal of Magnetism and Magnetic Materials*, *342*, (2013): 74-79.
[7]. Zhang, L. N., Hu, J. F., Chen, J. S., Ding, J., *Journal of Applied Physics*, *105*(7), (2009): 07A743.
[8]. Zhang, L. N., Hu, J. F., Chen, J. S., Ding, J., *Journal of Magnetism and Magnetic Materials*, *321*(17), (2009): 2643-2647.
[9]. Zangari, G., Lu, B., Laughlin, D. E., Lambeth, D. N., *Journal of Applied Physics*, *85*(8),(1999): 5759-5761.

[10]. Benaissa, M., Krishnan, K. M., Fullerton, E. E., Jiang, J. S., *IEEE Transactions on Magnetics*, *34*(4), (1998): 1204-1206.
[11]. Saravanan, P., Hsu, J. H., Reddy, G. L. N., Kumar, S., Kamat, S. V., *Journal of Alloys and Compounds*, *574*, (2013): 191-195.
[12]. Malhotra, S. S., Liu, Y., Shan, Z. S., Liou, S., Stafford, D. C., Sellmyer, D. J., *Journal of Applied Physics*,*79*(8), (1996): 5958-5960.
[13]. Verma, K. C., Kotnala, R. K., *Journal of Nanoparticle Research*, *13*(10), (2011): 4437-4444.
[14]. Schrefl, T., Fidler, J., Kronmüller, H.,*Physical Review B* 49.9 (1994): 6100.
[15]. Wyckoff, R. W. G., Crystal structures 1 (7–83). *American Mineralogist Crystal Structure Database*, (1963).
[16]. Singh, A., Neu, V., Tamm, R., Rao, K.S., Fahler, S., Skrotzki, W., Schultz, L. Holzapfel, B., *Applied Physics Letters*, *87*(7), 2005: 072505.
[17]. Zhang, L. N., Ding, J., Hue, J. F., Chen, J. S., *Nanoelectronics Conference, 2008. INEC 2008. 2nd IEEE International*, 1038-1042.
[18]. Patra, A. K., Neu, V., Fähler, S., Wendrock, H., Schultz, L., *Journal of applied physics*, *100*(4), (2006): 043905.
[19]. Zhang, J., Song, J. Z., Zhang, Y., Wang, F., Chen, B. C., Shen, B. G., Sun, J. R., *IEEE Transactions on Magnetics*, *47*(10), (2011): 2792-2795.
[20]. Goll, D., Seeger, M., Kronmüller, H., *Journal of Magnetism and Magnetic Materials*, *185*(1), (1998): 49-60.
[21]. Shao, Y. Z., Zhong, W. R., Lan, T., Lee, R. H., *International Journal of Modern Physics B*, *20*(01), (2006): 61-72.
[22]. Herzer, G., *Magnetics, IEEE Transactions on* 26.5 (1990): 1397-1402.
[23]. Fischer, R.,Schrefl, T., Kronmüller, H., Fidler, J., *Journal of Magnetism and Magnetic Materials*, *153*(1), (1996): 35-49.
[24]. Kronmüller, H., Fischer, R., Seeger, M., Zern, A.,*Journal of Physics D: Applied Physics*, *29*(9), (1996): 2274.