Interfacial Perpendicular Magnetic Anisotropy and Damping Parameter in Ultra Thin Co_2FeAl Films

Tim Mewes – University of Alabama

Citation of published version:

Cui, Y., et al. (2013): Interfacial Perpendicular Magnetic Anisotropy and Damping Parameter in Ultra Thin Co2FeAl Films, *Applied Physics Letters*, 102(16). https://doi.org/10.1063/1.4802952

APPLIED PHYSICS LETTERS **102**, 162403 (2013)

Interfacial perpendicular magnetic anisotropy and damping parameter in ultra thin Co_2FeAl films

Yishen Cui,[1] Behrouz Khodadadi,[2] Sebastian Schäfer,[2] Tim Mewes,[2] Jiwei Lu,[3] and Stuart A. Wolf[1,3]

[1]*Department of Physics, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Department of Physics and Astronomy, MINT Center, University of Alabama, Tuscaloosa, Alabama 35487, USA*
[3]*Department of Materials Science and Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*

(Received 13 March 2013; accepted 8 April 2013; published online 23 April 2013)

B2-ordered Co_2FeAl films were synthesized using an ion beam deposition tool. A high degree of chemical ordering ~81.2% with a low damping parameter (α) less than 0.004 was obtained in a 50 nm thick film via rapid thermal annealing at 600 °C. The perpendicular magnetic anisotropy (PMA) was optimized in ultra thin Co_2FeAl films annealed at 350 °C without an external magnetic field. The reduced thickness and annealing temperature to achieve PMA introduced extrinsic factors thus increasing α significantly. However, the observed damping of Co_2FeAl films was still lower than that of $Co_{60}Fe_{20}B_{20}$ films prepared at the same thickness and annealing temperature. © *2013 AIP Publishing LLC* [http://dx.doi.org/10.1063/1.4802952]

Recently, Spin Transfer Torque Random Access Memory (STT-RAM) has emerged as a potential universal memory.[1,2] In order to realize the scalability of STT-RAM with the same lithographic node transistor, a major challenge is to reduce the critical current density J_0 for free layer magnetization switching.[2] J_0 can be reduced by providing perpendicular magnetic anisotropy (PMA) in the ferromagnetic electrodes of magnetic tunnel junctions (p-MTJs).[2,3] Interfacial PMA due to the spin-orbit interaction (SOI) between Co/Fe and oxygen atoms has thus been adopted in p-MTJs by introducing interfaces such as CoFeB/MgO.[4–8] Additionally, free layer electrodes with a low damping parameter are also important for further reducing J_0.[9]

Co_2FeAl has been considered as a promising candidate for STT-RAM. In in-plane MTJs, it has been reported that by adopting Co_2FeAl as the free layer, a high tunnel magnetoresistance (TMR) has been achieved around 330%–340%.[10] A recent study has demonstrated PMA at the interfaces between the ultra thin Co_2FeAl layer and the MgO layer with conventional field annealing treatments at ~350 °C.[11] Additionally, Co_2FeAl films with a thickness ~50 nm shows an ultra low damping parameter ~0.001.[12] However, in Ref. 12, rapid thermal annealing (RTA) at 600 °C was required to achieve high chemical ordering and thus a low damping parameter in 50 nm thick Co_2FeAl. With an annealing treatment at 350 °C, a large damping parameter ~0.04 was reported in 10 nm thick Co_2FeAl films.[13] The annealing temperature must remain below 600 °C to maintain the interfacial PMA in Co_2FeAl,[11] while a decrease of the magnetic film thickness also leads to a sharp increase in the damping parameter.[7,14] A study of the damping parameter of ultra thin Co_2FeAl films with perpendicular magnetization is thus necessary to evaluate its practical advantage over CoFeB in p-MTJs and that is the essence of this work.

This letter reports the interfacial PMA and the damping parameter in ultrathin Co_2FeAl films. In the first step, we demonstrated the impact of the RTA temperature on the degree of the chemical ordering in thick Co_2FeAl films (~50 nm). The damping parameter was characterized in the sample with the highest ordering. Second, interfacial PMA was established in Co_2FeAl films with reduced thicknesses. The largest PMA was achieved with an annealing treatment at 350 °C for 10 s and no external magnetic field was applied during annealing. We will present the effects of film thickness and the annealing conditions on the damping parameter of the perpendicularly magnetized films, and also compare the damping parameters of the ultra thin Co_2FeAl films with those of $Co_{60}Fe_{20}B_{20}$ films.

All the films were synthesized using a Biased Target Ion Beam Deposition system (BTIBD).[15] The system has a base pressure below 7×10^{-8} Torr. The MgO (001) substrates were *in-situ* heated at 500 °C for 30 min and etched for 5 min with an argon/oxygen plasma to remove the surface contamination on the MgO substrates. A single stoichiometric Co_2FeAl target was used. For the structure and chemical ordering characterizations, Co_2FeAl of a thickness ~50 nm was deposited on the substrates and annealed in the RTA furnace at temperatures between 350 °C and 600 °C in vacuum (1.5×10^{-5} Torr). To establish PMA in Co_2FeAl ultra thin films, stacks of a structure Cr(40 nm)/Co_2FeAl(t)/MgO(2.3 nm)/Ru(5 nm) were deposited on the MgO substrates, where t is the effective thickness of Co_2FeAl. Cr was deposited as a seed layer at 200 °C. The XRD pattern indicated that highly crystalline Cr was epitaxially grown along MgO (001), and the AFM scan showed a smooth surface with a roughness below 0.15 nm within a scan range of 1 μm^2. MgO was synthesized by sputtering metallic Mg in an oxygen ambient. To prevent the over-oxidation of Co_2FeAl underneath the MgO layer, a thin Mg layer (0.7 nm) was inserted before the MgO deposition. To investigate the effect of annealing conditions on the PMA, ultra thin Co_2FeAl films were annealed at temperatures from 300 °C to 450 °C with the annealing time varied from 10 s to 300 s. No external magnetic field was applied during annealing treatments.

The film composition was $Co_{49.3}Fe_{25.4}Al_{25.2}$ determined by Energy-dispersive X-ray spectroscopy (EDX), and was close to the 50:25:25 ratio for the full Heusler alloy. The film crystal structure was characterized using a high resolution X-Ray diffractometer with Cu Kα radiation (Smart-lab®, Rigaku, Inc.), and the degree of chemical ordering was calculated accordingly. The magnetic behavior was characterized using a vibrating sample magnetometer (VSM) in a Quantum Design VersaLab. Ferromagnetic resonance (FMR) spectra were acquired using a broadband coplanar waveguide field-swept setup with frequencies up to 50 GHz. The spectra were fitted to determine the resonance field and the linewidth of the FMR.[16] The close-to-linear frequency dependence of the linewidth was subsequently used to estimate an effective damping parameter for the samples.[17]

Fig. 1(a) shows the 2θ scans of selected Co_2FeAl films (50 nm) annealed at temperatures 500 °C, 550 °C, and 600 °C. The diffraction peaks of the (002) and (004) from the Co_2FeAl were observed at 31.19° and 65.09°, respectively. A lattice parameter of 5.74 Å was obtained based on the peak positions, consistent with values reported previously.[18,19] The (004) peak is known as the fundamental peak as a result of the diffraction of the cubic crystal structure, while the (002) superlattice peak indicates the B2 chemical ordering in the film.[19] The degree of the B2 ordering S was estimated from the ratio of integrated peak intensities for (002) and (004).[20] S was improved with increasing annealing temperature from 350 °C to 600 °C as shown in Fig. 1(b). It reached 82.1% in Co_2FeAl films annealed at 600 °C. Correspondingly, the estimated damping parameter was less than 0.004, comparable to the value reported in the previous study.[12]

As the thickness of Co_2FeAl was reduced, perpendicular magnetic anisotropy was achieved due to the Co_2FeAl/MgO interfaces. The magnetic behavior was characterized in as-deposited samples with different Co_2FeAl thicknesses, as presented in Fig. 2. The effective thickness t varied from 0.57 nm to 1.90 nm excluding the magnetically "dead layer" ~0.9 nm.[15] The "dead" layer is inactive magnetically, and is likely due to the intermixing at the interface between Co_2FeAl and the Cr seeding layer. The extracted saturation magnetization of ~731 emu/cm^3 was obtained for the whole series. Fig. 2(a) shows the M-H loops of selected Co_2FeAl films measured in a perpendicular magnetic field, which indicates the crossover from in-plane anisotropy to perpendicular anisotropy as the thickness was reduced. The interfacial perpendicular anisotropy between a Co/Fe based ferromagnetic layer and a properly oxidized MgO layer has been well-explained using first-principles calculation. As a weak SOI is involved, hybridizations within Co/Fe-3d orbits and between Co/Fe-3d and O-2p orbits lift the degeneracy around the Fermi level. The energy level for out-of-plane orientation of magnetization is lowered; therefore, resulting in an interfacial perpendicular anisotropy.[4]

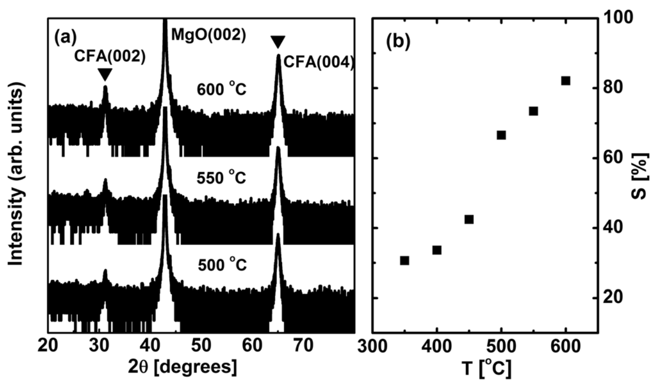

FIG. 1. (a) 2θ scans vs. the annealing temperature T for selected Co_2FeAl films (50 nm) annealed at 500 °C, 550 °C, and 600 °C, respectively. (b) The degree of the B2 ordering S vs. T ranged from 350 °C to 600 °C.

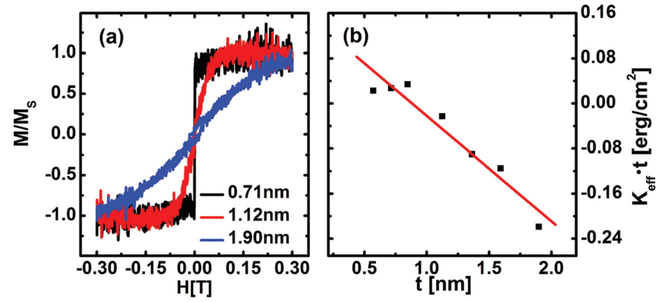

FIG. 2. (a) Out-of-plane hysteresis loops of the as-deposited Co_2FeAl films at room temperature. The selected effective thicknesses t are 0.71 nm (black), 1.12 nm (red), and 1.90 nm (blue). (b) $K_{eff} \cdot t$ vs. t for as-deposited Co_2FeAl films. The linear fitting is presented in red line.

The bulk and the interfacial contributions to the film anisotropy can be extracted from the phenomenological equation: $K_{eff} = K_V + K_S/t$, in which K_{eff} stands for the total effective anisotropy, K_V is the bulk anisotropy related to in-plane crystalline anisotropy and shape anisotropy while K_S is the surface anisotropy.[11,21] The total anisotropy becomes perpendicular as K_S/t overwhelms K_V. K_S can be estimated from the vertical axis intercept in the linear fitting between $K_{eff} \cdot t$ and t as shown in Fig. 2(b). In the as-deposited series, the surface anisotropy K_S was 0.16 erg/cm^2. It was increased to 0.65 erg/cm^2 via the RTA treatment at 350 °C, but slightly reduced to 0.64 erg/cm^2 after annealing at 400 °C.

The Co_2FeAl film thickness was fixed at 0.57 nm to investigate the impact of the annealing conditions on the total anisotropy K_{eff}. Fig. 3 shows the dependence of K_{eff} on the annealing temperature. As a baseline, K_{eff} was 0.38×10^6 erg/cm^3 in as-deposited films, and reached 1.9×10^6 erg/cm^3 at a temperature 350 °C, which is consistent with the value for Co_2FeAl/MgO reported in Ref. 11 and

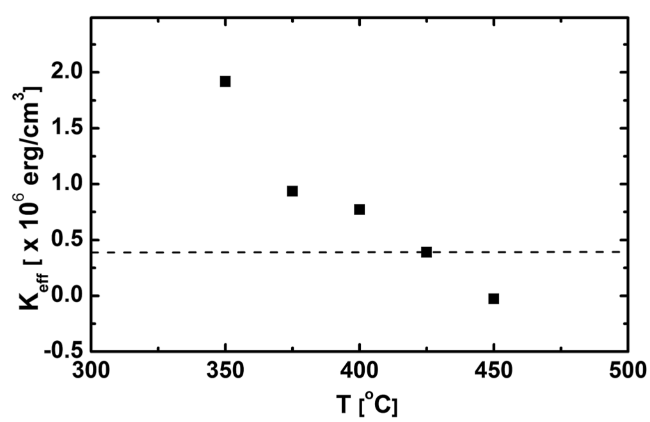

FIG. 3. Total film anisotropy (K_{eff}) as a function of the annealing temperature T in Co_2FeAl films (0.57 nm). The dashed line indicates the value of K_{eff} for the as-deposited sample. The magnetic anisotropy is perpendicular to the film with a positive K_{eff} while in-plane with a negative one.

comparable to that of other perpendicular anisotropy systems such as CoFeB/MgO (Ref. 7) and CoFeGe/MgO.[22] At 450 °C, K_{eff} changed orientation from out-of-plane to in-plane. In the as-deposited sample, both Co_2FeAl and MgO layers were amorphous so that there was a lack of long range ordering at the interface. In addition, presumably the protective Mg layer was naturally oxidized and oxygen interacted with Co/Fe atoms forming a Co/Fe oxide at the Co_2FeAl/MgO interfaces. Excess oxygen atoms induced a local charge redistribution eliminating the energy splitting for orbits Co/Fe-d_{z2} and O-p_z thus suppressing the PMA.[4–6] After annealing treatments, Co/Fe oxide was likely reduced while the crystallinity was improved at the interfaces leading to an increasing PMA. When the temperature exceeded 350 °C, the interfacial PMA started deteriorating possibly due to the reduction of proper Co/Fe-O bonding at the Co_2FeAl/MgO interfaces.[6] This was also evident when we studied the impact of annealing time on K_{eff} keeping the annealing temperature at 400 °C. The annealing time of ∼10 s yielded a perpendicular magnetization with K_{eff} of 0.8×10^6 erg/cm^3. When the annealing time exceeded 30 s, the in-plane magnetic anisotropy exceeded the interfacial PMA.

Fig. 4 shows the damping parameters for films of Co_2FeAl and $Co_{60}Fe_{20}B_{20}$ after annealing at 350 °C and 400 °C. $Co_{60}Fe_{20}B_{20}$ samples have the same stack structure as Co_2FeAl samples. The arrow highlights the Co_2FeAl sample with a perpendicular magnetization. The damping parameter for the perpendicularly magnetized sample was 0.012, much higher than the value observed in thick Co_2FeAl films. The increased damping parameter could be attributed to the effects from the decreased thickness and the reduced annealing temperature. With the annealing temperature fixed, the damping parameter was decreased from 0.012 to 0.009 as the Co_2FeAl thickness increased from 1.36 nm to 1.90 nm. A similar trend was also observed in the $Co_{60}Fe_{20}B_{20}$ reference samples. The thickness effect has been reported in the CoFeB system previously and possible origins were discussed.[7,14] On the other hand, the damping parameter decreased from 0.01 to 0.009 as the annealing temperature increased from 350 °C to 400 °C for films with a thickness of 1.90 nm. The decrease of the damping parameter might be due to the suppression of surface defects and the improvement of the B2 chemical ordering with an elevated annealing temperature. It was indicated that a high B2 chemical ordering was crucial in achieving a low damping parameter in Co_2FeAl (Ref. 12) and other ordered alloys.[9,23] As demonstrated previously in this letter, the B2 ordering can be improved by raising the annealing temperature. However, annealing treatments above 425 °C were not compatible with achieving the interfacial PMA. The B2 chemical ordering was sacrificed to some extent, leading to an increased damping parameter. Additionally, it is worth noting that through a conventional furnace annealing treatment at 350 °C, an increased damping parameter ∼0.04 was reported in a structure of MgO(001)/Cr(40 nm)/Co_2FeAl(10 nm).[13] The authors ascribed it to the residual magnetic moments in the Cr and the diffusion of Cr atoms into the Co_2FeAl layer.[13] In our work, the rapid thermal annealing at the same temperature yielded a damping parameter of ∼0.01 in MgO(001)/Cr(40 nm)/Co_2FeAl(1.9 nm)/MgO (2.3 nm), which might suggest that the diffusion of Cr was significantly reduced likely due to the very short annealing time (∼10 s) based on the assumption of Cr diffusion.

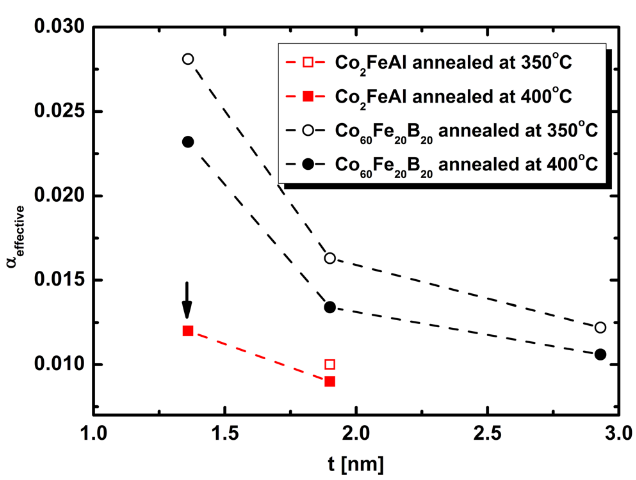

FIG. 4. Effective damping parameters α of Co_2FeAl and $Co_{60}Fe_{20}B_{20}$ films as a function of the effective thickness t. The open (solid) red squares represent Co_2FeAl films annealed at 350 °C (400 °C), and the open (solid) black circles represent $Co_{60}Fe_{20}B_{20}$ annealed at 350 °C (400 °C). The arrow highlights the Co_2FeAl sample with perpendicular magnetization.

The comparison of the damping parameters between Co_2FeAl and $Co_{60}Fe_{20}B_{20}$ samples shows that those obtained in Co_2FeAl samples were still lower than that of those corresponding $Co_{60}Fe_{20}B_{20}$ samples with the same stack structure and the same annealing treatment (Fig. 4).[24] Based on our results, the Heusler alloy Co_2FeAl would still have the advantage of a lower damping parameter than $Co_{60}Fe_{20}B_{20}$, important for lowering the critical current density for spin torque switching. The improvement, however, may be less than expected due to the enhanced damping parameter in thin Co_2FeAl films with interfacial PMA.

In summary, we obtained B2 ordered Co_2FeAl films (50 nm) via RTA treatments and demonstrated the dependence of the degree of B2 ordering on the annealing temperature. A damping parameter below 0.004 was obtained in the sample with the highest B2 ordering. As the Co_2FeAl thickness was reduced, interfacial PMA was established and improved by applying RTA treatments without the assistance of an external magnetic field. The annealing temperature was optimized at 350 °C to maximize the interfacial PMA, which was comparable to previously reported value for Co_2FeAl/MgO (annealed in a magnetic field) and other systems with interfacial PMA such as CoFeB and CoFeGe. The reduction of the film thickness and annealing temperature led to an increased damping parameter of ∼0.012 in the perpendicularly magnetized Co_2FeAl film. On the other hand, the damping parameters of Co_2FeAl films were still lower compared to those of $Co_{60}Fe_{20}B_{20}$ films synthesized in the same fashion. Ultra thin Co_2FeAl films could serve as an alternative material to $Co_{60}Fe_{20}B_{20}$ for reducing the critical current density in STT-RAMs, though a trade-off between the large interfacial PMA and the low damping parameter has to be taken into account in perpendicular STT-RAM applications.

S.A.W., J.W.L., and Y.C. acknowledge the financial supports from DARPA (Award No. HR0011-09-C-0023) and

NSF Grant (Award No. ECCS-0948138). T.M. and B.K. are supported by NSF-CAREER Award (Award No. 0952929).

[1]S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[2]Y. Huai, AAPPS Bull. **18**, 33 (2008).
[3]H. Yoda, T. Kishi, T. Nagase, M. Yoshikawa, K. Nishiyama, E. Kitagawa, T. Daibou, M. Amano, N. Shimomura, S. Takahashi *et al.*, Curr. Appl. Phys. **10**, e87 (2010).
[4]H. X. Yang, M. Chshiev, B. Dieny, J. H. Lee, A. Manchon, and K. H. Shin, Phys. Rev. B **84**, 054401 (2011).
[5]R. Shimabukuro, K. Nakamura, T. Akiyama, and T. Ito, Physica E **42**, 1014 (2010).
[6]W. G. Wang, S. Hageman, M. Li, S. Huang, X. Kou, X. Fan, J. Q. Xiao, and C. L. Chien, Appl. Phys. Lett. **99**, 102502 (2011).
[7]S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, and H. Ohno, Nature Mater. **9**, 721 (2010).
[8]W. X. Wang, Y. Yang, H. Naganuma, Y. Ando, R. C. Yu, and X. F. Han, Appl. Phys. Lett. **99**, 012502 (2011).
[9]H. Lee, Y.-H. A. Wang, C. K. A. Mewes, W. H. Butler, T. Mewes, S. Maat, B. York, M. J. Carey, and J. R. Childress, Appl. Phys. Lett. **95**, 082502 (2009).
[10]W. Wang, E. Liu, M. Kodzuka, H. Sukegawa, M. Wojcik, E. Jedryka, G. H. Wu, K. Inomata, S. Mitani, and K. Hono, Phys. Rev. B **81**, 140402(R) (2010).
[11]Z. Wen, H. Sukegawa, S. Mitani, and K. Inomata, Appl. Phys. Lett. **98**, 242507 (2011).
[12]S. Mizukami, D. Watanabe, M. Oogane, Y. Ando, Y. Miura, M. Shirai, and T. Miyazaki, J. Appl. Phys. **105**, 07D306 (2009).
[13]H. Sukegawa, Z. Wen, K. Kondou, S. Kasai, S. Mitani, and K. Inomata, Appl. Phys. Lett. **100**, 182403 (2012).
[14]X. Liu, W. Zhang, M. J. Carter, and G. Xiao, J. Appl. Phys. **110**, 033910 (2011).
[15]Y. Cui, W. Yin, W. Chen, J. Lu, and S. A. Wolf, J. Appl. Phys. **110**, 103909 (2011).
[16]S. Keshavarz, Y. Xu, S. Hrdy, C. Lemley, T. Mewes, and Y. Bao, IEEE Trans. Magn. **46**, 1541 (2010).
[17]B. Heinrich, J. F. Cochran, and R. Hasegawa, J. Appl. Phys. **57**, 3690 (1985); T. Mewes, R. L. Stamps, H. Lee, E. Edwards, M. Bradford, C. K. A. Mewes, Z. Tadisina, and S. Gupta, IEEE Magn. Lett. **1**, 3500204 (2010).
[18]K. H. J. Buschow and P. G. van Engen, J. Magn. Magn. Mater. **25**, 90 (1981).
[19]S. Okamura, A. Miyazaki, N. Tezuka, S. Sugimoto, and K. Inomata, Mater. Trans. **47**, 15 (2006).
[20]K. Barmak, J. Kim, L. H. Lewis, K. R. Coffey, M. F. Toney, A. J. Kellock, and J.-U. Thiele, J. Appl. Phys. **98**, 033904 (2005); H. Bremers, J. Hesse, H. Ahlers, J. Sievert, and D. Zachmann, J. Alloys Compd. **366**, 67 (2004).
[21]M. T. Johnsony, P. J. H. Bloemenzx, F. J. A. den Broedery, and J. J. de Vriesz, Rep. Prog. Phys. **59**, 1409 (1996).
[22]M. Ding and S. J. Poon, Appl. Phys. Lett. **101**, 122408 (2012).
[23]C. Liu, C. K. A. Mewes, M. Chshiev, T. Mewes, and W. H. Butler, Appl. Phys. Lett. **95**, 022509 (2009).
[24]Lower damping parameters have been reported in ultra thin $Co_{40}Fe_{40}B_{20}$, which cannot be directly compared to our results given the differences in the composition and the capping layers. [M. Oogane, T. Wakitani, S. Yakata, R. Yilgin, Y. Ando, A. Sakuma, and T. Miyazaki, Jpn. J. Appl. Phys., Part 1 **45**, 3889 (2006); C. L. Wang, S. H. Huang, C. H. Lai, W. C. Chen, S. Y. Yang, K. H. Shen, and H. Y. Bor, J. Phys. D: Appl. Phys. **42**, 115006 (2009).]