AIP | Applied Physics Letters

FeNi-based magnetoimpedance multilayers: Tailoring of the softness by magnetic spacers

A. V. Svalov, E. Fernandez, A. Garcia-Arribas, J. Alonso, M. L. Fdez-Gubieda, and G. V. Kurlyandskaya

Citation: Applied Physics Letters **100**, 162410 (2012); doi: 10.1063/1.4704984
View online: http://dx.doi.org/10.1063/1.4704984
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/100/16?ver=pdfcov
Published by the AIP Publishing

FeNi-based magnetoimpedance multilayers: Tailoring of the softness by magnetic spacers

A. V. Svalov,[1,2] E. Fernandez,[1] A. Garcia-Arribas,[1] J. Alonso,[1] M. L. Fdez-Gubieda,[1] and G. V. Kurlyandskaya[1,a)]
[1]*Departamento de Electricidad y Electrónica, Universidad del País Vasco, 48080 Bilbao, Spain*
[2]*Institute of Physics and Applied Mathematics, Ural Federal University, Ekaterinburg, 620002 Russia*

(Received 23 February 2012; accepted 4 April 2012; published online 20 April 2012)

The microstructure and magnetic properties of sputtered permalloy films and FeNi(170 nm)/X/FeNi(170 nm) (X = Co, Fe, Gd, Gd-Co) sandwiches were studied. Laminating of the thick FeNi film with various spacers was done in order to control the magnetic softness of FeNi-based multilayers. In contrast to the Co and Fe spacers, Gd and Gd-Co magnetic spacers improved the softness of the FeNi/X/FeNi sandwiches. The magnetoimpedance responses were measured for $[FeNi/Ti(6\,nm)]_2/FeNi$ and $[FeNi/Gd(2\,nm)]_2/FeNi$ multilayers in a frequency range of 1–500 MHz: for all frequencies under consideration the highest magnetoimpedance variation was observed for $[FeNi/Gd(2\,nm)]_2/FeNi$ multilayers. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4704984]

Magnetic softness is an important issue for thin films as a material for magnetic sensors. However, it is not easy to achieve adequate softness in FeNi films prepared by sputtering when their thickness is greater than the critical thickness. This is the so-called "transcritical" state of thin magnetic films characterized by the presence of an out-of-plane magnetization component, stripe domains and increased coercivity.[1,2] The perpendicular anisotropy responsible for the formation of the "transcritical" state in FeNi films prepared by sputtering might be a consequence of strain-caused magnetostriction and/or the columnar structure of the films.[3] The critical thickness value (t_c) usually corresponds to the interval of 100 to 350 nm and strongly depends on deposition conditions and characteristics of the sputtering equipment.[3–9] One way to tackle this problem is to laminate the thick FeNi film with thin nonmagnetic spacers, while keeping the individual FeNi layer thickness below the critical thickness corresponding to a transition into a "transcritical" state. For example, Ag,[10] Ta and Cr,[4] Cu,[11] Ti,[12] and Mo (Ref. 9) have been used as the spacer materials. Limited studies were done for magnetic spacers in case the materials of the spacer and the magnetic layer had different crystalline structure.[13] It was shown that this interrupted the growth of principal layers and preserved magnetic coupling throughout the multilayer structure.

It should also be noted that the lamination by a nonmagnetic spacer may further improve the softness of the multilayer structure. In case of strong magnetostatic interactions between the domain walls situated in different magnetic layers, these domain walls form structures with a partially closed magnetic flux. Thus, the reduced energy of paired domain walls diminishes the coercivity in multilayer films.[14] However, the dependence of the coercive force (H_c) of two-layer magnetic films on the thickness of a nonmagnetic spacer (L_X) is non-monotonic: H_c falls rapidly with the L_X increase, but after $L_X \sim 1$ nm the coercive force rises again.[15] It was shown that the pairs of the domain walls with low magnetostatic energy can be formed with a higher probability in the case of a positive coupling over the interface between the layers.[14,15] For nonmagnetic spacers, this positive coupling may be due to magnetostatic coupling such as "orange rind"[16] and magnetostatic coupling through "magnetization ripple."[17] With the increase of the nonmagnetic spacer thickness, the magnetic interaction becomes gradually weaker resulting in the appearance of non-doubled walls, a rise of the average wall energy, and an increase of H_c. Thus, the mechanisms of positive coupling between layers together with the magnetostatic interaction of the domain walls mainly determine the hysteretic properties of the sandwiches.[15] One can therefore expect that magnetic spacers, when used for laminating the thick FeNi film, can lead to an additional advantage. On one hand, the magnetic spacer can interrupt the growth of FeNi layers and prevent the transition into a "transcritical" state if the spacer has a different lattice constant or crystalline structure. On the other hand, an appropriate magnetic interaction between FeNi layers separated by magnetic spacers can be an instrument to obtain a less pronounced minimum in the $H_c(L_X)$ dependence.

In this work, we studied magnetic properties of permalloy films with different types of spacers prepared by dc magnetron sputtering in order to gain insight on the role of different spacers in the formation of magnetically soft FeNi multilayers and their magnetoimpedance (MI). We chose Co, Fe, Gd, and amorphous $Gd_{21}Co_{79}$ alloy as the magnetic spacers, which have a crystallography different from that of the fcc structure of the permalloy. In addition, their Curie temperatures (T_C) and hence the interaction exchange constants were quite different.

The samples were grown onto glass substrates at room temperature. The background pressure was 3×10^{-7} mbar. Deposition was performed in an Ar atmosphere with 3.8×10^{-3} mbar pressure. The deposition rates were previously calibrated as 26 nm/min for FeNi layers and 1 nm/min for all spacer materials. The thickness of the FeNi layers was 170 nm taking into account the fact that under these deposition conditions the transition into the "transcritical" state takes place at the thickness of about 200 nm.[8] The thickness of spacers (L_X) was varied in the range of 0.5 to 20 nm.

a)Electronic mail: galina@we.lc.ehu.es.

A constant magnetic field of 250 Oe was applied parallel to the film plane in order to induce a uniaxial magnetic anisotropy.

Small angle x-ray scattering in grazing incidence (GISAXS) measurements were carried out at the BM16 beamline of the ESRF (Grenoble) in order to obtain additional information about the structure of the FeNi films. The hysteresis loops were recorded by means of the magneto-optical Kerr effect (MOKE). For magnetoimpedance measurements, the MI elements were deposited with a metallic mask in the shape of elongated strips of 0.5 mm × 10 mm. The MI of these samples (total impedance (Z) change in a magnetic field) was measured as a function of the external magnetic field for a frequency (f) range of 1 to 500 MHz. The system was calibrated in order to extract the internal values of the impedance from the total measured signals by using the method and experimental system described in Ref. 18. The MI ratio was defined with respect to the sample saturated in the maximum applied field $H = 150$ Oe as follows: $\Delta Z/Z(H) = 100 \times (Z(H)\text{-}(Z(H = 150\,\text{Oe}))/Z(H = 150\,\text{Oe})$.

Fig. 1 shows the 2D GISAXS pattern obtained for a FeNi thin film. In the reciprocal space, q_y and q_z correspond to the directions parallel and perpendicular to the surface, respectively. The most relevant feature of the scattering pattern is the presence of a well defined anisotropy with two elongated intensity maxima appearing at both sides of the beam-stopper. This can be associated with a columnar growth of the FeNi nanograins inside the thin film.[19,20] This result suggests that the perpendicular anisotropy responsible for the formation of "transcritical" state might have resulted from the columnar thin-film microstructure.

There was a difference in the influence of the selected spacers on magnetic properties of FeNi(170 nm)/X/FeNi(170 nm) samples. "Transcritical" shape of hysteresis loops remained for any thickness of the cobalt and iron. For example, Figs. 2(a) and 2(b) show two loops for Co spacers. A similar situation was observed for Fe spacers (not sown here). In the case of Gd-Co a "transcritical" state disappears under a spacer thickness above 4 nm (Figs. 2(e) and 2(f)), the minimum coercivity ($H_c \sim 0.2$ Oe) corresponded to a Gd-Co layer thickness of ~7–9 nm (Fig. 3(b)). For Gd the "transcritical" shape loop disappears under a spacer thickness above 1 nm (Figs. 2(c) and 2(d)), the minimum coercivity ($H_c \sim 0.1$ Oe) corresponded to the $L_{Gd} \sim 2$–3 nm (Fig. 3(a)).

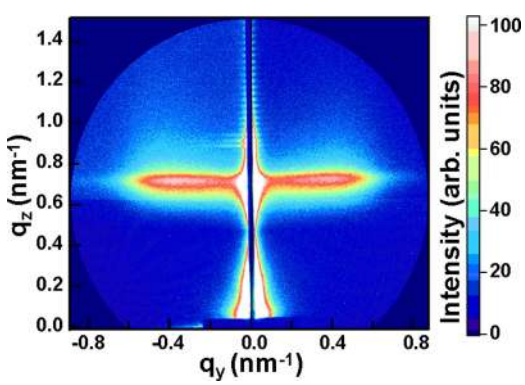

FIG. 1. GISAXS pattern obtained for a FeNi film.

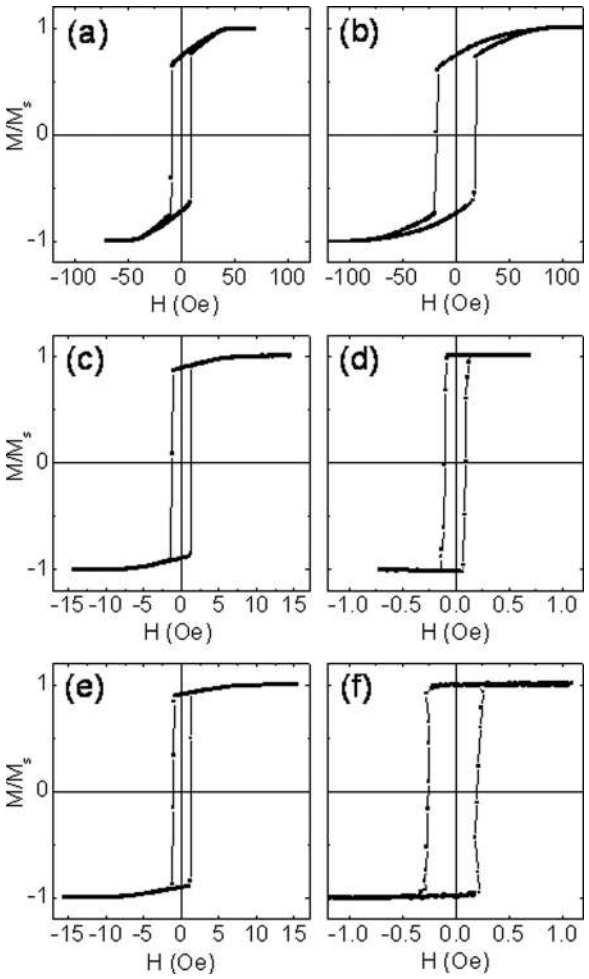

FIG. 2. MOKE hysteresis loops for FeNi/X(nm)FeNi samples, where X: (a) Co(5 nm), (b) Co(20 nm), (c) Gd(1 nm), (d) Gd(2 nm), (e) Gd-Co(4 nm), and (f) Gd-Co(6 nm).

The important question in a connection with the presence of the "transcritical" state in FeNi/(Co, Fe)/FeNi sandwiches is about the crystalline structure of Co and Fe spacers. The equilibrium crystalline phase of bulk cobalt at room temperature is hcp. Ultra thin Co films can have a fcc structure when grown on a substrate with the fcc structure under certain growth conditions. Generally, the fcc FeNi layer can act as a template for fcc growth of the cobalt layer. At the same time, the fcc phase of cobalt is metastable at room-temperature and as the film thickness grows beyond a few monolayers, the transition from the fcc stacking to the hcp structure occurs gradually as the film thickness increases.[21] A similar situation occurs for Fe spacers. Ultra thin Fe films can have a metastable fcc structure when grown onto substrate with a fcc structure, but in the coverage range near 10 monolayers, a structural transition from fcc to stable bcc proceeds in the film with an increasing thickness.[22] Therefore, we assume that in our case, Co and Fe spacers have hcp and bcc structures, respectively, when its thickness increases beyond few nanometers. Thus, Co and Fe spacers interrupt the growth of fcc FeNi layers, while the "transcritical" state is preserved in FeNi/(Co, Fe)/FeNi sandwiches.

The above mentioned result does not coincide with data from earlier work[13] where it was shown that laminating the

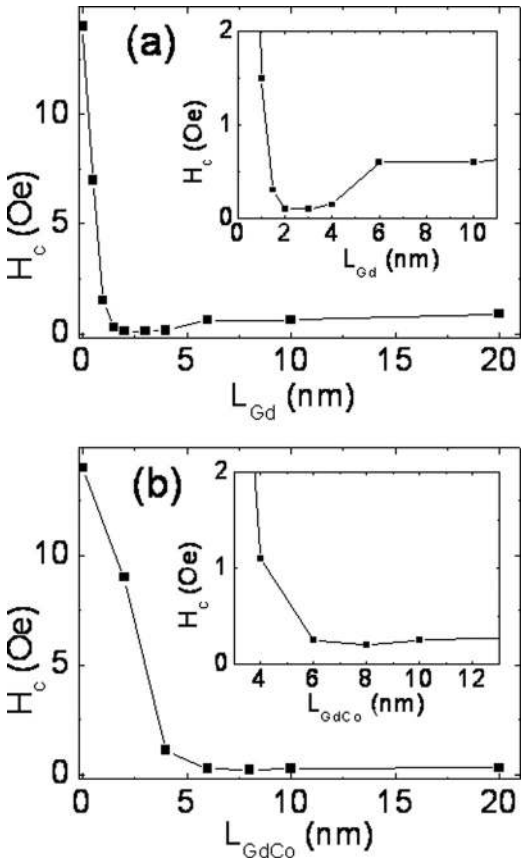

FIG. 3. Dependence of the coercivity of FeNi/Gd/FeNi (a) and FeNi/Gd-Co/FeNi (b) samples on the spacer thickness. Insets show the same graph in a smaller scale.

thick fcc $Ni_{77}Fe_{14}Cu_5Mo_4$ film with thin bcc CoFe magnetic spacers eliminates the "transcritical" shape of a hysteresis loop and restores the magnetic softness of this material. One can suppose that this discrepancy is caused by differences in the mechanism of formation of perpendicular magnetic anisotropy in the films. In the above mentioned work,[13] it was shown that in $Ni_{77}Fe_{14}Cu_5Mo_4$ film the effective perpendicular magnetic anisotropy was determined by the stress through the magnetostriction, which increased with film thickness. Laminating this film with layers having different lattice constants or crystal structures reduces stress as large as 200-fold. This means that the perpendicular anisotropy constant (K_p) for the $Ni_{77}Fe_{14}Cu_5Mo_4$ film decreases multiply as well because $K_p \sim 3/2\ \lambda \cdot \sigma$, where λ is the magnetostriction constant and σ is the stress. Thus, spacer introduction significantly increases the t_c value due to $t_c \sim 1/K_p^{3/2}$ [Ref. 28]. Therefore, in this case, interlayering with thin bcc CoFe spacers allows to obtain a multilayer film with continuous magnetic coupling and a large t_c value.

In FeNi films studied here, perpendicular anisotropy results from the columnar structure of the films and K_p depends weakly on the film thickness. Therefore laminating the thick FeNi film with spacers of different lattice constants or crystal structures interrupts growth of FeNi, but almost does not affect K_p. Nonmagnetic spacers "mechanically" divide thick FeNi film into magnetically separate layers with thicknesses less than t_c. By contrast, magnetic spacers from strong magnets like Co or Fe preserve powerful magnetic coupling between FeNi layers and "effective" t_c values of these layered structures are not significantly different from t_c for FeNi single layer film. In this way, the total thickness of this hard magnetically coupled structure exceeds the t_c value and "transcritical" state takes place in FeNi/(Co, Fe)/FeNi sandwiches.

The T_C of amorphous $Gd_{21}Co_{79}$ alloy is above room temperature, but appreciably smaller than the T_C of iron and cobalt films. Its own value of exchange coupling is less than the interaction exchange constants of Co and Fe. Therefore FeNi layers are coupled weaker by means of Gd-Co spacer comparing with Co or Fe spacers. As a result, at the thickness of the Gd-Co spacer above 4 nm, the FeNi layers in FeNi/(Gd-Co)/FeNi sandwiches behave more likely as separated layers and the thickness of each magnetic layer does not exceed the t_c of FeNi single layer film. It is worth to mention that Gd-Co alloy is a ferrimagnet with a compensation temperature (T_{comp}) slightly above the room temperature (about 310 K). Selected measurements of the hysteresis loop of the samples with Gd-Co spacer were done both above and below the T_{comp}. These hysteresis loops had practically identical shape. Therefore it is logical to conclude that in the case under consideration the main parameter of the Gd-Co spacer is the value of the exchange interaction constant but not the type of the magnetic sublattice (Gd or Co) predominant at a given temperature.

The "transcritical" hysteresis loops were not observed for hcp Gd interlayers with a thickness above 1 nm. However, there is an uncertainty about magnetic state of Gd spacers. The T_C value for bulk Gd is 293 K. In addition, for thin Gd films at L_{Gd} below 5 nm the T_C decreases sharply.[23,24] This is a reason to consider Gd spacers as nonmagnetic spacers. On the other hand, the proximity of Gd and FeNi layers contributes to an enhancement of the Gd magnetization at FeNi/Gd interfaces at temperatures above the T_C of Gd thin film. This magnetization is induced by a strong antiferromagnetic exchange interaction with the magnetically ordered FeNi layers.[25] The interlayer exchange between FeNi and Gd decays exponentially from the interfaces to the centre of the Gd spaces,[26] but Gd spacers of few nanometers can be highly magnetized.[27] Furthermore, cooling of the FeNi/Gd(6 nm)FeNi sample down to 190 K, i.e., much lower than the T_C of Gd(6 nm) layer,[24] does not change the hysteresis loop shape shown in Fig. 2(f). Therefore, one can conclude that the introduction of *magnetic* Gd spacers eliminates the "transcritical" state in FeNi/Gd/FeNi sandwiches. The T_C and exchange constant of Gd film is less than one of the amorphous $Gd_{21}Co_{79}$ alloy and the disappearance of the "transcritical" loops at a smaller spacer thickness in the Gd case is understandable.

Thus, in terms of preventing an occurrence of the "transcritical" state, the magnetic properties, namely, the interaction exchange constant of spacer materials, are no less important than the structural features.

From the viewpoint of providing additional magnetic softness, the Gd and Gd-Co spacers act differently. For the single layer FeNi(170 nm) film, $H_c = 0.7$ Oe. The Gd-Co alloy allows us to have H_c less then the mentioned value for FeNi/Gd-Co/FeNi sandwiches with a spacer thickness of up to 20 nm (Figs. 3(b)). In the case of Gd spacers above

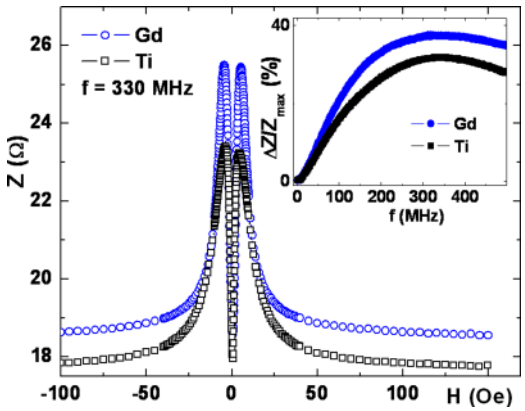

FIG. 4. Field dependence of the total impedance of $[FeNi/Ti(6\,nm)]_2/FeNi$ and $[FeNi/Gd(2\,nm)]_2/FeNi$ multilayers. Inset shows the frequency dependence of the maximum value $\Delta Z/Z_{max}$ of the MI ratio.

$L_{Gd} > 3$ nm, the coercive force of FeNi/Gd/FeNi sandwiches rises rapidly and again reaches the level of coercivity of the FeNi(170 nm) film at a thickness spacer above 5 nm. But among the investigated magnetic spacers, the best magnetic softness was obtained for Gd: at $L_{Gd} > \sim 2$–3 nm $H_c = 0.1$ Oe. This H_c value is similar to H_c for the Ti spacer with a thickness of 6 nm in FeNi/Ti multilayers.[12]

It is interesting to compare soft magnetic FeNi/Ti and FeNi/Gd multilayers as a material for MI sensors. Fig. 4 shows the frequency dependence of the maximum of the MI ratio and total impedance dependence on the external field for both $[FeNi/Ti(6\,nm)]_2/FeNi$ and $[FeNi/Gd(2\,nm)]_2/FeNi$ multilayers. One can see that for all frequencies under consideration $\Delta Z/Z(H)$ is higher for the $[FeNi/Gd]_2/FeNi$ multilayers. The absolute maxima appear at a frequency of about 330 MHz in both cases and which is why we selected this frequency for the analysis of the field dependence. The impedance variation in the field close to the anisotropy field of the multilayer is much higher for $[FeNi/Gd]_2/FeNi$ multilayers. More studies are necessary in order to explain the sharp increase of the MI value in the Gd containing multilayers, but the difference in the mechanisms of magnetic interaction between the FeNi layers is probably a consequence: for $[FeNi/Gd]_2/FeNi$ multilayers, a direct exchange via Gd spacers is possible, but for $[FeNi/Ti(6\,nm)]_2/FeNi$ layered structures the "orange peel" and "magnetization ripple" magnetostatic mechanisms should be essential. In MI configuration, the application of the external field can significantly change the magnetic state of the multilayered structure because of the complex dynamic of the magnetic moments.

In conclusion, structural and magnetic properties of FeNi(170 nm)/X/FeNi(170 nm) sandwiches (X = Co, Fe, Gd, Gd-Co) were studied. The perpendicular magnetic anisotropy responsible for the formation of the "transcritical" state in thick FeNi films is likely a result of the columnar thin-film microstructure. In order to avoid the transition into a "transcritical" state, we laminatied the thick FeNi film with various magnetic spacers. The Co and Fe spacers of any thickness did not prevent an appearance of the out-of-plane magnetization component in FeNi-based sandwiches. The Gd and amorphous Gd-Co spacers not only eliminate the "transcritical" state, but even enhance the sandwich magnetic softness. The coercivity minimum (~ 0.1 Oe) was achieved for the Gd spacers with the thickness of 2 nm. The MI ratio measured for $[FeNi/Ti(6\,nm)]_2/FeNi$ and $[FeNi/Gd(2\,nm)]_2/FeNi$ multilayers is higher in the last case which is most likely a consequence of the difference in the mechanisms of magnetic interaction between the FeNi layers.

This work was supported by RFBR-11-02-00288-a, URFU-215, MEC-MAT2011-27573-C04 and G°V° IT-347-07 grants. Magnetic measurements were performed at SGIker service of UPV-EHU. We thank I. Orue, F. Fauth and K. Wickersham for excellent support. BM16 beamline time (ESRF, Grenoble) is specially acknowledged.

[1]M. Coïsson, F. Celegato, E. Olivetti, P. Tiberto, F. Vinai, and M. Baricco, J. Appl. Phys. **104**, 033902 (2008).
[2]A. V. Svalov, G. V. Kurlyandskaya, H. Hammer, P. A. Savin, and O. I. Tutynina, Tech. Phys. **49**, 868 (2004).
[3]N. Amos, R. Fernández, R. Ikkawi, B. Lee, A. Lavrenov, A. Krichevsky, D. Litvinov, and S. Khizroev, J. Appl. Phys. **103**, 07E732, (2008).
[4]S. F. Cheng, P. Lubitz, Y. Zheng, and A. S. Edelstein, J. Magn. Magn. Mater. **282**, 109 (2004).
[5]J. Ben Youssef, N. Vukadinovic, D. Billet, and M. Labrune, Phys. Rev. B **69**, 174402 (2004).
[6]C. A. Ramos, E. Vassallo Brigneti, J. Gómez, and A. Butera, Physica B **404**, 2784 (2009).
[7]T. Dastagir, W. Xu, S. Sinha, H. Wu, Y. Cao, and H. Yua, Appl. Phys. Lett. **97**, 162506 (2010).
[8]A. V. Svalov, R. Aseguinolaza, A. Garcia-Arribas, I. Orue, J. M. Barandiaran, J. Alonso, M. L. Fdez-Gubieda, and G. V. Kurlyandskaya, IEEE Trans. Magn. **46**, 333 (2010).
[9]M. Romera, R. Ranchal, D. Ciudad, M. Maicas, and C. Aroca. J. Appl. Phys. **110**, 083910 (2011).
[10]R. L. Sommer, A. Gündel, and C. L. Chien, J. Appl. Phys. **86**, 1057 (1999).
[11]G. V. Kurlyandskaya, L. Elbaile, F. Alves, B. Ahamada, R Barrue, A. V. Svalov, and V. O. Vas'kovskiy, J. Phys.: Condens. Matter. **16**, 6561 (2004).
[12]G. V. Kurlyandskaya, A. V. Svalov, E. Fernandez, A. Garcia-Arribas, and J. M. Barandiaran, J. Appl. Phys. **107**, 09C502 (2010).
[13]W. F. Egelhoff, Jr., J.Bonevich, P.Pong, C. R.Beauchamp, G. R.Stafford, J.Unguris, and R. D.McMichael, J. Appl. Phys., **105**, 013921 (2009).
[14]E. Feldtkeller. J. Appl. Phys. **39**, 1181 (1968).
[15]V. O. Vas'kovskii, P. A. Savin, V. N. Lepalovskii, and A. A. Ryazantsev, Phys. Solid State **39**, 1958 (1997).
[16]L. Neel, Comptes Rendus **255**, 1676 (1962).
[17]H. Hoffmann, IEEE Trans. Magn. **4**, 32 (1968).
[18]D. de Cos, J. M. Barandiaran, A. Garcia-Arribas, V.O. Vas'kovskiy, and G. V. Kurlyandskaya, IEEE Trans. Magn. **44**, 3863 (2008).
[19]D. Babonneau, F. Petroff, J. L. Maurice, F. Vaurès, and A. Naudon, Appl. Phys. Lett. **76**, 2892 (2000).
[20]D. Babonneau, F. Pailloux, J. P. Eymery, M. F. Denanot, Ph. Guérin, E. Fonda, and O. Lyon, Phys. Rev. B **71**, 035430 (2005).
[21]B.P. Tonner, Z.-L. Han, and J. Zhang Phys. Rev. B **47**, 9723 (1993).
[22]P. Schmailzl, K. Schmidt, P. Bayer, R. Döll, and K. Heinz, Surf. Sci. **312**, 73 (1994).
[23]M. Farle, K. Baberschke, U. Stetter, A. Aspelmeier, and F. Gerhardter, Phys. Rev. B **47**, 11571 (1993).
[24]A. V. Svalov, V. O. Vas'kovskiy, J. M. Barandiarán, K. G. Balymov, A. N. Sorokin, I. Orue, A. Larrañaga, N. N. Schegoleva, and G. V. Kurlyandskaya, Solid State Phenom. **168–169**, 281 (2011).
[25]D. Haskel, G. Srajer, J. C. Lang, J. Pollmann, C. S. Nelson, J. S. Jiang, and S. D. Bader. Phys. Rev. Lett. **87**, 207201 (2001).
[26]A.V. Svalov, J. M. Barandiaran, V. O. Vas'kovskiy, G. V. Kurlyandskaya, L. Lezama, N. G. Bebenin, J. Gutierrez, and D. Schmool, J. Alloys Compd. **327**, 5 (2001).
[27]J. L. Prieto, M. G. Blamire, and J. E. Evetts, Phys. Rev. Lett. **90**, 027201 (2003).
[28]Y. Sugita, H. Fujiwara, and T. Sato, Appl. Phys. Lett. **10**, 229 (1967).