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# Exchange Bias in FeMn/M (M = FeNi, Gd, Tb) Films

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**Abstract**

Microstructure and hysteretic properties of magnetic multilayers were studied for $Fe_{50}Mn_{50}$/M structures, where M = $Fe_{19}Ni_{81}$, Gd or Tb. Comparative analysis of the hysteresis loops measured for the temperature range 5÷350 K showed that Gd is not involved in the interlayer exchange coupling with antiferromagnetic $Fe_{50}Mn_{50}$ layer, while in the case of Tb definite indications of such interaction were observed. It is assumed that a qualitative difference in the magnetic behavior of these rare earth layers can be caused by differences in their structural features.


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*Keywords:* exchange bias; hysteresis properties; multilayers; rare earth; X-ray diffraction.

## 1. Introdution

Sensor devises is important application of nanostructured magnetic materials (Dieny, 2004). It is focused on functional magnetic materials with enhanced sensitivity. In some cases the presence of the unidirectional magnetic anisotropy (exchange bias) is a key point of specialized electronic device. This anisotropy is most often implemented in soft magnetic layers of multilayer films that are in the exchange coupling with antiferromagnetic layers. It manifests itself in the shifting of the hysteresis loop along the magnetic field axis. A typical example of such films is $Fe_{19}Ni_{81}$/$Fe_{50}Mn_{50}$ bilayers (Giri et al., 2011) or more complex film structures based on them (Chen et al., 2007). The uniformity of the crystal structure of layered elements is very important in this case (Kim et al., 2009). At the same time, magnetic properties of the multilayers containing $Fe_{50}Mn_{50}$ antiferromagnetic layer in contact with the

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layers of rare earth metals have barely been investigated. The specific of the electronic structure of these magnets unlikely allows the realization of spin-dependent scattering of the conduction electrons in their case. However, the indirect exchange interaction leads to a tendency of formation of complex magnetic structures in rare-earth metals and their alloys. This feature together with the relatively low temperatures of magnetic ordering is the reason for the search of new functional properties that can be implemented by varying the magnetic field and temperature in exchange biased layered structures containing rare earth layers. This work is devoted to the comparative study of the features of the exchange bias with 3d- and 4f-ferromagnetic layers in the multilayered structures containing Fe50Mn50 antiferromagnetic layers. As representatives of the group of rare earth metals Gd and Tb were selected, because they have the largest values of the Curie temperature and differ from each other from the point of view of the features of magnetic anisotropy.

## 2. Samples preparation

Multilayered structures were deposited onto Corning glass substrates at ambient temperature by Orion 8 sputtering installation with five sputter sources. Background pressure was as high as $3 \times 10^{-7}$ mbar and the argon pressure during deposition was as high as $2 \times 10^{-3}$ mbar. Multilayere films were formed by sequential sputtering of homogeneous targets of pure metals (Gd, Tb) and alloys (FeNi, FeMn) in the presence of high-frequency bias voltage applied to a substrate. A uniform magnetic field of 250 Oe oriented parallel to the substrate plane was also applied during deposition. Electrical bias promoted the formation of homogeneous microstructure and technological magnetic field specified the direction of the easy magnetization in the plane of the films (Gorkovenko et al., 2014). A power of 200 W was applied to the target. Thicknesses of the layers were adjusted by setting the sputtering time according to the deposition rates obtained previously by using Dektak-150 instrument. The deposition rates were in the interval of 0.05 to 0.1 nm/sec for different materials. Samples had following multilayered structure: $SiO_2$/Ta(5)/Fe19Ni81(5 nm)/Fe50Mn50(20 nm)/M(40 nm)/Ta(5 nm). M means one of the metals: Fe19Ni81 (sample 1), Gd (sample 2), Tb (sample 3).

In multilayered structures, auxiliary Ta buffer layer and directly following Fe19Ni81 adjacent layers with the same thickness of 5 nm played an important role in the microstructure formation. They created necessary conditions for the formation of fcc crystalline structure in the Fe50Mn50 layer. It is considered that the probability for fcc crystalline structure formation is higher for the deposition of Fe50Mn50 layer onto the surface of the layer with fcc structure, for example onto the surface of permalloy layer. This fcc structure is required for formation of antiferromagnetic properties of FeMn. (Chen et al., 2007). In addition, a contributing factor to enhance the magnetic bias, in particular, due to the formation of crystalline texture, is the presence of a buffer layer of Ta, separating permalloy from the substrate (Kim et al., 2009). Outer Ta layer plays a protective function.

The crystalline structure of the films was studied by X-ray diffraction (XRD) using Philips X'PertPRO diffractometer with the $Cu_{K\alpha}$ radiation source. The magnetic properties of the samples were measured by MPMS XL7 EC magnetometer in the 5÷350 K temperature range.

## 3. Results

### 3.1. Structure

Figure 1 shows the normalized diffractograms of the studied films. The main common feature observed in all samples is the presence of the bright peak near $2\theta = 43.5^{o}$ angle, which was identified as a (111) peak of fcc crystal lattice of FeMn. The intensity of the (111) reflection was used for the normalization procedure for the intensity in the each diffractogram. Note that the reflections from the other Fe50Mn50 crystal planes in the 2θ range of 10 to 90 degrees were absent. This indicates the high level of the crystallographic texturing of FeMn layers. For the Fe50Mn50 layers in all samples, quantitative analysis of the XRD profile gives the same lattice parameter values (a = 0.360 nm) and crystallite size (13-18 nm) found by the Scherer formula (Patterson, 1939). It allows us to suggest that magnetic properties of these layers are similar. Their main feature is an antiferromagnetic ordering.

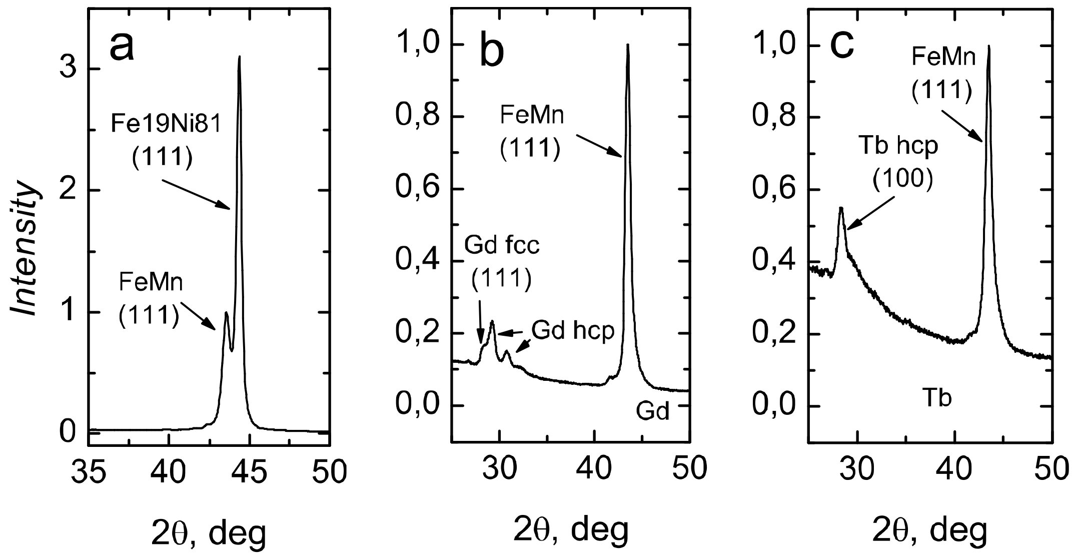


Fig.1. XRD spectra of Fe50Mn50/M multilayers with different M-layers: (a) - Fe19Ni81; (b) - Gd; (c) - Tb.

Other peaks that were present in the XRD spectra after analysis have been attributed to M-layers. They indicated a significant difference in the structural state of permalloy and rare earth metals. The relatively narrow and intense line near $2\theta = 44.3^{o}$ for the sample 1 (Fig.1, a) was associated with fcc crystal lattice (a = 0.354 nm) of permalloy. It indicates the presence of a strong crystalline (111) texture in permalloy layer. The average size of the crystallites in this layer is about 35 nm. The XRD spectrum for sample 2 (Fig.1, b) near the $2\theta = 30^{o}$ has few relatively weak peaks that can be attributed to cubic and hexagonal phases of Gd. For the sample 3 only one diffraction peak near $2\theta = 30^{o}$ was observed. It can be attributed to Tb hexagonal crystal lattice. Estimation of the average crystallite size yields a value of about 13 nm for both rare earth metals.

*3.2. Magnetic properties*

The main objective of this work was to investigate the presence and effectiveness of exchange coupling between the antiferromagnetic Fe50Mn50 layer and rare earth metals layers that are significantly differ from each other in the magnetic anisotropy features (Vas'kovskiy et al., 2015). The main results were obtained from the analysis of hysteresis loops $m(H)$, measured along the easy magnetization axis at different temperatures $T$. Comparative evaluation of the features of such hysteresis loops for the samples 1 and 2 with M = Fe19Ni81 and M = Gd are shown in Fig. 2. It can be seen that at room temperature (Fig.2, a) the shape of the hysteresis loop for sample 1 is a stepped one. It is a superposition of hysteresis loops of the main layer and seed layer of permalloy. Both minor loops are shifted along the magnetic field axis that indicates the presence of exchange coupling between ferromagnetic and antiferromagnetic layers. Quantitative characteristics of interlayer interaction can be given through the exchange bias field, which values constitute $H_{e1}$ and $H_{e2}$ for permalloy layers with thicknesses of 40 and 5 nm, respectively. With $T$ decrease, exchange bias field both layers significantly increases. This is evidenced by a change of the hysteresis loops (compare Fig.2, a, c) and the dependence $H_{e1}(T)$, $H_{e2}(T)$, as shown in Fig. 3. Magnetic moments of the different layers estimated from the corresponding minor hysteresis loops at $T < 300$ K showed weak temperature dependence (less than 10% for the interval under consideration). Thus, the temperature variation of the energy of interaction for these layers with the magnetic field is small. This suggests that the main reason for temperature changes in the exchange bias field is variation of exchange stiffness and magnetic anisotropy of the antiferromagnetic layer.

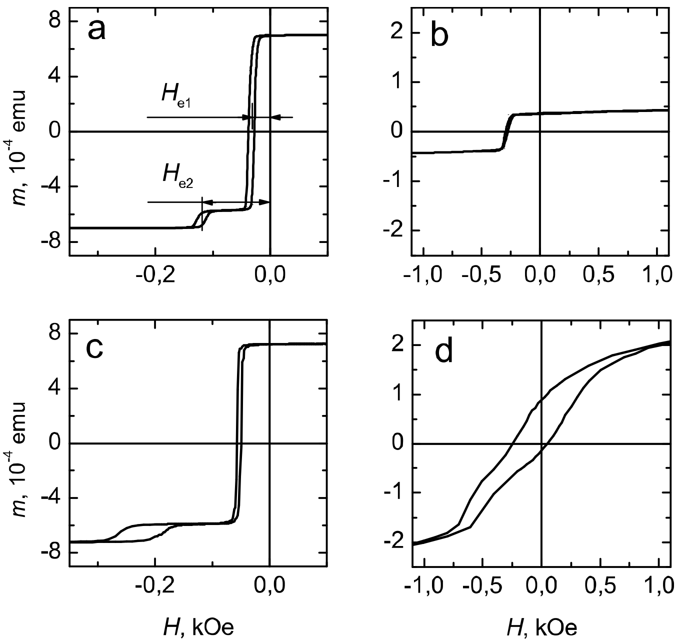


Fig.2. Hysteresis loops of the Fe50Mn50/M multilayers with M = Fe19Ni81 (a, c) и M = Gd (b, d), measured at different temperatures: a, b - 300; c, d - 150 K.

For the sample 2 at room temperature only single-stage magnetization reversal was observed (Fig.2, b). The value of this magnetization jump testifies that this magnetization reversal is connected only with the seed permalloy layer, but the main Gd layer state is a paramagnetic. For this layer, magnetic ordering occurs at lower temperatures in the range 250-300 K. This is reflected in the growth of the amplitude of the magnetization reversal and the emergence of non-linear non-hysteretic plots in the *m*(*H*) dependences. They describe almost reversible Gd magnetization near the Curie temperature. A further decrease of temperature leads to an intensive increase of the magnetic hysteresis in the rare earth metal layer and the transformation of general hysteresis loop shape to the shape shown in Fig.2, d. The distortions presented in the loop, reflect the fact of overlapping of minor loops of the two ferromagnetic layers. At first glance it seems that both loops are shifted along the magnetic field axis as in the case of sample 1. However, our estimates show that the behaviour is different.

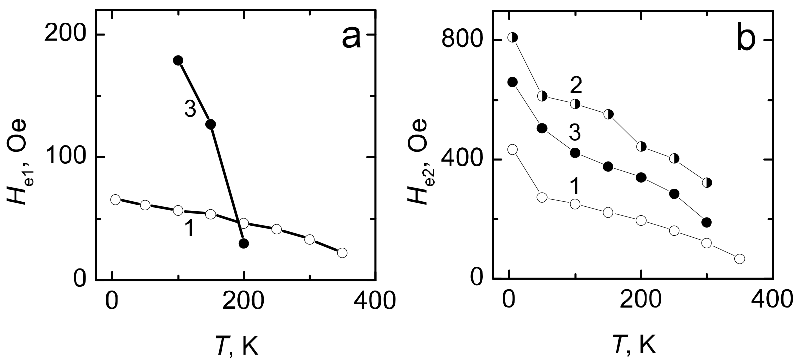


Fig.3. Temperature dependencies of exchange bias fields $H_{e1}$ (a) и $H_{e2}$ (b) of the Fe50Mn50/M multilayers with different layers M: Fe19Ni81 - curves 1; Gd - curve 2; Tb - curves 3.

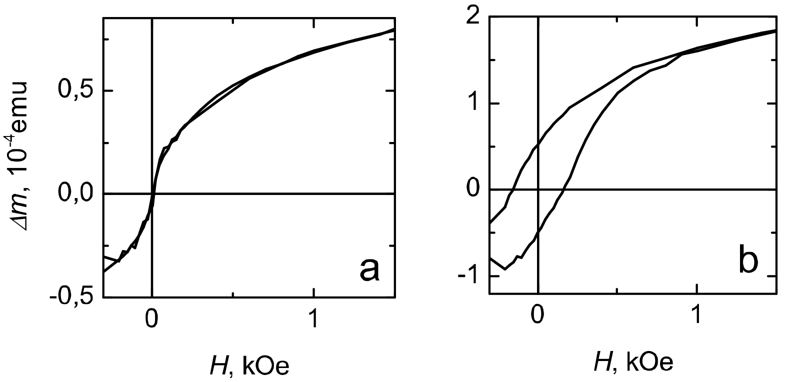


Fig.4. Selected parts of difference between the hysteresis loops $\Delta m(H) = m_T(H) - m_{300}(H)$ for sample with M = Gd at temperatures of 250 K (a) and 150 K (b)

Similar result were obtained at the other temperatures, except T < 50 K, where the coercive force of the seed permalloy layer was too large for the correct application of this technique. It follows that most likely Gd layer had no exchange bias, and therefore the exchange interaction between the Gd and Fe50Mn50 layer was absent. This result is in agreement with results published by Hossu et al. (2008), where the Gd/FeMn interface did not induce exchange bias in [FeMn10nm/Gd4nm/(Co4nm/Gd4nm)4/FeMn10nm] multilayer. However the Fe50Mn50 layer creates exchange bias for permalloy seed layer. Moreover, the $H_{e2}$ field value for the sample 2 is much larger than one for sample 1 (Fig.3, b). This fact can be considered as indirect evidence of the absence of interlayer exchange coupling between the Gd and Fe50Mn50 layers. Lower $H_{e2}$ values for the sample 1 were caused apparently by the perturbing effect which exerted on the magnetic structure of the antiferromagnetic layer due to the magnetization reversal of the main permalloy layer. Since Gd layer has not the interlayer exchange coupling with Fe50Mn50 layer therefore Gd layer does not performed the appropriate action.

This result seems to be unusual, since it is known that in heterogeneous film structures, containing layers of Gd and ferromagnetic 3d-metals or their alloys, there is a sufficiently strong interlayer exchange interaction (Vas'kovskiy et al. (1999), Nagura et al. (2002), Altuncevahir et al. (2003), Kravtsov et al. (2009), Ranchal et al. (2012), Barth et al. (2008)). This gives us a basis to consider a different reason for the absence of the exchange bias in Gd layer. Hypothetically, it can be assumed that it is the relative long-range exchange of rare earth metals, through which Gd can participate in an exchange interaction with both magnetic sub-systems of antiferromagnetic Fe50Mn50. Thus, the resulting biasing effect can be hidden. This situation unlikely seems to be a reality, because if so it should lead to a strong magnetic hysteresis in Gd layer.

Sample 3 contains Tb layer and at room temperature demonstrates magnetic properties similar to ones of the sample 2. His hysteresis loop is formed by the seed permalloy layer because the rare earth metal is a paramagnetic state (Fig.5, a). But in the low-temperature range Tb makes a significant contribution to the process of the sample magnetization reversal. It consists of the appearance in hysteresis loops the high-field parts with relatively high magnetic susceptibility. Initially (T = 200 K), they were non-hysteretic (Fig.5, b), but with a decrease of the temperature change of the magnetic moment on the loop become irreversible. As a result, the hysteresis loop acquire quite characteristic shape (Fig.5, c) which includes almost rectangular loop of the permalloy seed layer and rather narrow inclined loop of the Tb main layer. It can be seen, that the general shape, is significantly differ from the hysteresis loop of the sample 2. The reason for such a behavior is a local magnetic anisotropy of the Tb crystallites (Nikitin, 1989).

Analysis of interlayer coupling in the sample 3, as in the previous case, was performed by allocation of selected parts of minor hysteresis loops Tb layer. The only important difference was following: $m_{250}(H)$ dependence was taken as a reference loop, i.e. the hysteresis loop measured at T = 250 K. It is caused by that the magnetic ordering of Tb which occurs at lower temperatures than temperatures for magnetic ordering of Gd. A typical dependence of $\Delta m(H)$ for the sample 3 is shown in Fig.5, d. As can be seen, that it shifted along the magnetic field axis, perhaps be

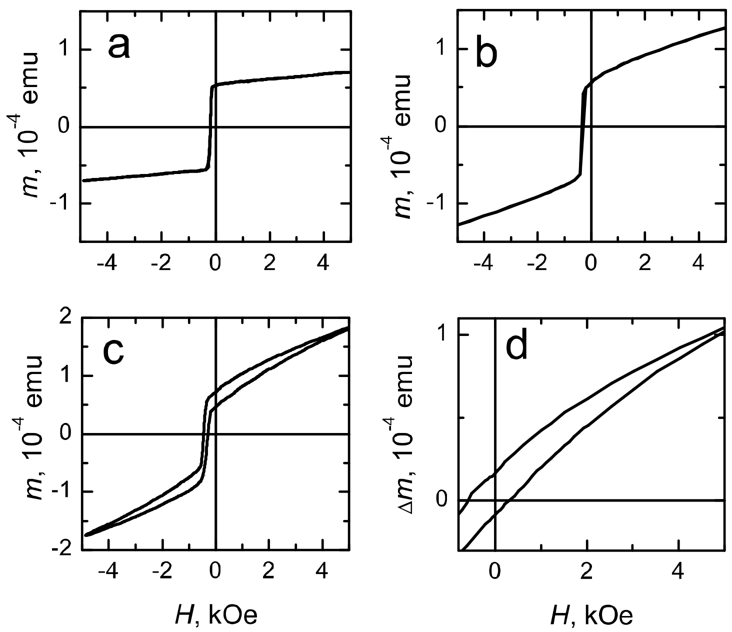


Fig.5. Experimental (a, b, c) and calculated (d) hysteresis loops of the sample 3 with M = Tb at different temperatures: a - 300; b - 200; c - 150; d - 150 K.

due to the exchange coupling between ferromagnetic and antiferromagnetic layers. Fig.3, a (curve 3) shows selected values of bias field $H_{e1}$ of Tb layer with sharp increase of the magnetic bias with decreasing of the temperature. However, the obtained results do not allow us to make clear conclusion about the origin of this shift, whether it is due to the interlayer exchange coupling. Another possible reason for the magnetic bias can be a strong magnetic hysteresis of Tb layer. Certain magnetic prehistory can also lead to a bias of the minor hysteresis loop. To resolve this indeterminacy additional research is needed, including, for example, the magnetization reversal of samples in sufficiently strong magnetic fields.

An argument in a favor of exchange nature of magnetic bias is related to the value of $H_{e2}$ field of the sample 3 (Fig.3, b). It is higher than in the sample with permalloy, but lower than in the sample with Gd. The last circumstance can be interpreted as the result of exchange effects of layer Tb on the antiferromagnetic layer. In this case, the question arises about the reason for the qualitative difference of Tb and Gd as the parts involved into the interlayer interaction with antiferromagnetic Fe50Mn50. It is possible that it is the consequence of the difference in the structural state of rare earth metals, and in particular, structural non-homogeneities and multiphase structure of Gd observed in the investigated samples.

## 4. Conclusions

Microstructure, and the interlayer interaction in magnetic multilayers with contacting ferromagnetic Fe19Ni81, Gd or Tb layers, and antiferromagnetic Fe50Mn50 layer were investigated. Obtained experimental data and modeling results confirm the established point of view on the presence of the effective exchange coupling in the Fe19Ni81/Fe50Mn50 structure and do not show it in Gd/Fe50Mn50 multilayered structure. In the Tb/Fe50Mn50 structure the observed magnetic bias of rare earth metal layer most likely caused by the indicated interlayer exchange coupling. It is assumed that the distinction between the different rare earth metals in the ability to take part in the interlayer exchange coupling can be due to their structural features, and in particular, non-homogeneities and multiphase structure of Gd.

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## References

Dieny, B., 2004. Magnetoelectronics. In: Johnson, M. (Ed.). Elsevier, Amsterdam, pp. 67–149.

Giri, S., Patra, M., Majumdar, S., 2011. Exchange bias effect in alloys and compounds. J. Phys.: Condens. Matter 23, 073201.

Chen, K.-C., Wu, Y. H., Wu, K.-M., Wu, J. C., Horng, L., 2007. Effect of annealing temperature on exchange coupling in NiFe/FeMn and FeMn/NiFe systems. J. Appl. Phys.101, 09E516.

Kim, K.-Y., Choi, H.-C., You, C.-Y., Lee, J.-S., 2009. Exchange bias and compositional depth profiles of annealed NiFe/FeMn/CoFe trilayers. J. Appl. Phys. 105, 07D715.

Gor'kovenko, A.N., Lepalovskij, V.N., Vas'kovskiy, V.O., Savin, P.A., Shchegoleva N.N., 2014. Effect of RF substrate bias on exchange coupling in Fe20Ni80/Fe50Mn50 films. Solid State Phenomena 215, 278-283.

Patterson, A.L., 1939 The Scherrer formula for X-Ray particle size determination. Phys. Rev. 56, 978-982.

Vas'kovskiy, V.O., Adanakova, O.A., Balymov, K.G., Kulesh, N.A., Svalov, A.V., Stepanova, E.A., 2015. Specific features of the formation of atomic magnetic moments in amorphous films RE-Co (RE = La, Gd, Tb). Phys. Solid State. 57, 1142-1147.

Hossu, M.R., Demirtas, S., Salamon, M.B., Koymen, A.R., 2008. Exchange bias in a ferrimagnetic/antiferromagnetic system. 2008 APS March Meeting, 53, http://meetings.aps.org/Meeting/MAR08/Session/X32.8.

Vas'kovskiy, V.O., Svalov, A.V., Vázquez, M., Hernando, A., Kurlyandskaya, G.V., García, D., Gorbunov, A.V., 1999. Magnetic anisotropy peculiarities of Gd/Co films near the magnetic compensation state. J. Magn. Magn. Mater. 203, 295-297.

Nagura, H., Takanashi, K., Mitani, S., Saito, K., Shima, T., 2002 Current-perpendicular-to-plane magnetoresistance in Co/Gd multilayers with twisted spin structure. J. Magn. Magn. Mater. 240, 183–185.

Altuncevahir, B., Koymen, A.R., 2003. Positive and negative exchange bias in CoNi/Gd/CoNi trilayers and CoNi/Gd bilayers. J. Magn. Magn. Mater. 261, 424-432.

Kravtsov, E., Haskel, D., te Velthuis, S. G. E., Jiang, J. S., Kirby, B. J., 2009. Complementary polarized neutron and resonant X-ray magnetic reflectometry measurements in Fe/Gd heterostructures: Case of inhomogeneous intralayer magnetic structure. Phys. Rev. B. 79 (2009), 134438.

Ranchal, R., Choi, Y., Romera, M., Freeland, J. W., Prieto, J. L., Haskel, D., 2012. Influence of the Fe content on the Gd magnetic ordering temperature in Ni1−xFex/Gd multilayers. Phys. Rev. B. 85, 024403.

Barth, A., Treubel, F., Marszałek, M., Evenson, W., Hellwig, O., Borschel, C., Albrecht, M., Schatz, G., 2008. Magnetic coupling in Gd/Ni bilayers. J. Phys.: Condens. Matter. 20, 395232-6.

Nikitin, C.A., 1989. Magnetic properties of rare earth metals and their alloys (in Russian), Moscow State University, Moscow.