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# Biquadratic interlayer interaction in three-layer CoNi/Si/FeNi films

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The paper presents the results of experimental studies using the electron magnetic resonance method of three-layer CoNi/Si/FeNi films, where one layer is magnetically hard and the other is magnetically soft. It was found that the introduction of a non-magnetic semiconductor silicon interlayer significantly affects the interlayer interaction. When studying the angular dependences of the resonance fields related to different subsystems, a direct observation of the biquadratic interlayer interaction was obtained, which value depends both on the thickness of the silicon interlayer and on the measurement temperature.

**Keywords:** magnetic resonance, exchange interaction, three-layer films, biquadratic interaction.

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Magnetic nanoscale multilayer film systems with different alternating layers command attention both because of the great variety of phenomena observed in them and because of possible practical applications. A great part of devices and appliances using magnetic layered structures operate based on the effects of spin-dependent transport. In this regard, the structures where materials with different physical properties, such as magnetics and semiconductor [1], are interfaced are promising. The efficiency of transport properties control is determined by interlayer interactions and the state of interfaces between different layers. Significant progress has been made in understanding the mechanisms of interlayer interactions and their role in magnet-dependent transport [2]. Since the creation of multilayer magnetic films with nanoscale layer thicknesses, various mechanisms of interlayer interactions have been established [3]. One type of interaction was found to be biquadratic interlayer interaction, which in the first experiments [4] by magneto-static and magneto-optical methods was recorded by the presence of perpendicular domain structure in Fe/Cr/Fe films. It was found [5] that in a certain interval of chromium interlayer thicknesses, the biquadratic interaction exceeds the bilinear interaction. In superlattices $[Fe/Cr]_n$ using the magnetic resonance method, it was found [6] that the specific features of the resonance spectrum behavior are explained when biquadratic exchange and fourth-order magnetic anisotropy are taken into account. The microwave giant magnetoresistive effect was found, and it was analyzed taking into account bilinear and biquadratic contributions to the interlayer interaction energy [7]. Also in structures with wedge-shaped chromium interlayer, the temperature-dependent behavior of both bilinear and biquadratic interaction [8,9] was observed by methods of magnetostatics, magneto-optical Kerr effect and Mandelstam–Brillouin light scattering. The same type of interaction is implemented in Fe/$Me$/Fe ($Me$ = Al, Au) [9] films, with a rapid decrease in the biquadratic interaction constant with increasing temperature [10] in films with a small thickness of Al interlayer. In superlattices $[Fe/FeSi]_n$ [11] and structures with a semiconductor interlayer Fe/Si/Fe [12] a temperature-dependent biquadratic interaction was also observed.

In our case, the choice of the research object was determined by the fact that the CoNi/Si/FeNi film structures have a number of interesting properties. First, the coexistence of the effects of magnetic spring and exchange bias [13] was found earlier, and second, the presence of a semiconductor interlayer suggests temperature- and/or magneto-dependent interlayer interactions.

The films were sputtered onto a glass substrate on an ultrahigh vacuum magnetron sputtering machine from „Omicron“ (with system for controlling film thickness during growth) at a base pressure of $10^{-10}$ Torr. The nickel content in the CoNi layer was 19.5 at.%, and in the FeNi layer — 83 at.%. The thickness of the CoNi layer was $t_h = 53$ nm and was chosen based on the considerations that it should exhibit the properties of a magnetically hard layer when measured in accessible magnetic fields. The soft magnetic FeNi layer was $t_s = 72$ nm thick. The thicknesses used for the non-magnetic semiconductor layer were: $t_{Si} = 2.4$ and 3.3 nm. When sputtering the CoNi layer, the substrate temperature was 450 K for better adhesion with the substrate and to induce uniaxial anisotropy, and then the substrate was cooled to 373 K. The silicon layer and permalloy layer were sputtered at this substrate temperature to eliminate (minimize) silicide formation. The layer deposition rate $V \approx 0.15$ nm/s. To induce the magnetic anisotropy axis, sputtering and subsequent cooling were carried out in a magnetic field ($\sim 16$ kA/m). The ratio of magnetic anisotropy values of magnetically hard and soft magnetic materials at room temperature is about two orders of magnitude and only increases as the temperature decreases. The angle determining the direction of the

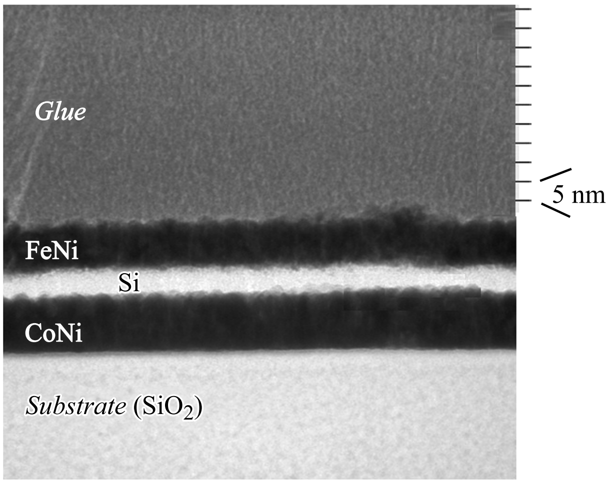


**Figure 1.** Cross section of the CoNi/Si/FeNi film.

external magnetic field was counted from the easy direction of magnetization in the film plane. The thickness of the layers was determined by X-ray spectroscopy. Electron-microscopic studies of the cross section were carried out using a JEOL JEM-2100 electron microscope (during sample preparation on a Gatan PIPS unit). No traces of silicide phases were found. It was also determined that the CoNi film was polycrystalline and was in hexagonal phase. The electron magnetic resonance spectra were measured on a Bruker E 580 CW EPR spectrometer operating at a microwave frequency of 9.48 GHz.

Fig. 1 shows a cross-sectional image of the CoNi/Si/FeNi film, where the thicknesses of the magnetic layers and silicon are made comparable for clarity. It can be seen that the interfaces between the silicon layer and both magnetic layers are clear, and there is no indication that there is a transition metal-silicon alloy.

Magnetic resonance measurements of single layers show that the magnetic moment lies in the film plane, and uniaxial induced anisotropy occurs in the film plane. The anisotropy fields at room temperature obtained from electron magnetic resonance data are of the order of $\sim 20$ kA/m for CoNi and $\leq 100$ A/m for CoNi and $\leq 100$ A/m for FeNi, which approximately coincides with the coercivity [13]. In the case of the CoNi/FeNi bilayer structure, ferromagnetic interaction is implemented between the layers.

The situation changes dramatically when a non-magnetic silicon interlayer is introduced between the magnetically hard layer (CoNi) and the soft magnetic layer (FeNi). If the magnetic energies of the individual layers are much larger than the interlayer interaction energy, then the observed resonance absorption peaks can be genealogically attributed to the CoNi and FeNi layers. We denote these peaks as low-field (*1*) and high-field (*2*), respectively. In the three-layer structure, the location of the resonance peaks ($H_r$) inherent in different magnetic layers is significantly shifted. At small silicon thicknesses ($t_{\mathrm{Si}} = 2.4$ nm) the peak assigned to the CoNi layer shifts to the zero-field region (curve *1* in Fig. 2, *a*), due to which it is impossible to extract the resonance absorption parameters. When the thickness of the silicon interlayer is increased ($t_{\mathrm{Si}} = 3.3$ nm) both peaks are clearly observed (Fig. 2, *b*).

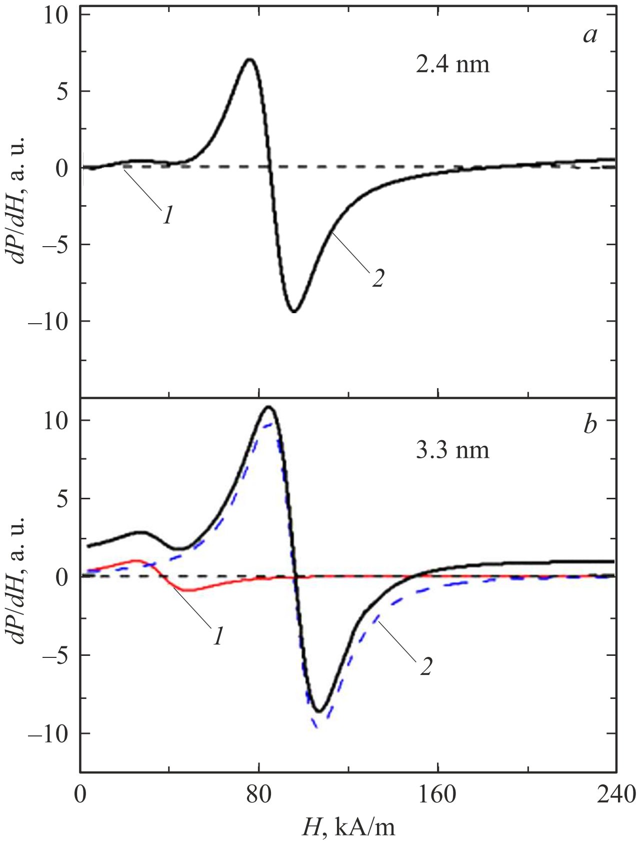


**Figure 2.** Resonance spectrum of CoNi/Si/FeNi films. $t_{\mathrm{Si}} = 2.4$ (*a*) and 3.3 nm (*b*). *1, 2* — peaks assigned to CoNi and FeNi layers, respectively. The solid line refers to the experiment. $T = 10$ K.

When studying the angular dependences of the resonance fields, it was found that the curves corresponding to the CoNi and FeNi layers within the structure behave differently when the direction of the external magnetic field in the film plane is changed (Fig. 3). It was found that for the film with $t_{\mathrm{Si}} = 2.4$ nm the microwave absorption line from the CoNi layer appears at $T > 150$ K, and at higher temperatures both peaks have the same angular dependence (Fig. 3, *a* and *c*). For the film with $t_{\mathrm{Si}} = 3.3$ nm both lines exist in the entire temperature range, but the angular dependences for the resonance peaks of each layer are transformed as the temperature increases (Fig. 3, *b* and *d*): the minimum of the resonance field of the peak *1* at low temperatures falls at the angle $\varphi = 0$, whereas the peak *2* at this angle has a maximum value $H_r$. This means that the magnetizations of CoNi and FeNi layers have perpendicular equilibrium directions. As the temperature is further increased, the peak of the FeNi subsystem is shifted relative to the CoNi peak.

It is well-known [14] that the magnetic in-plane anisotropy field of FeNi has a magnitude not more than

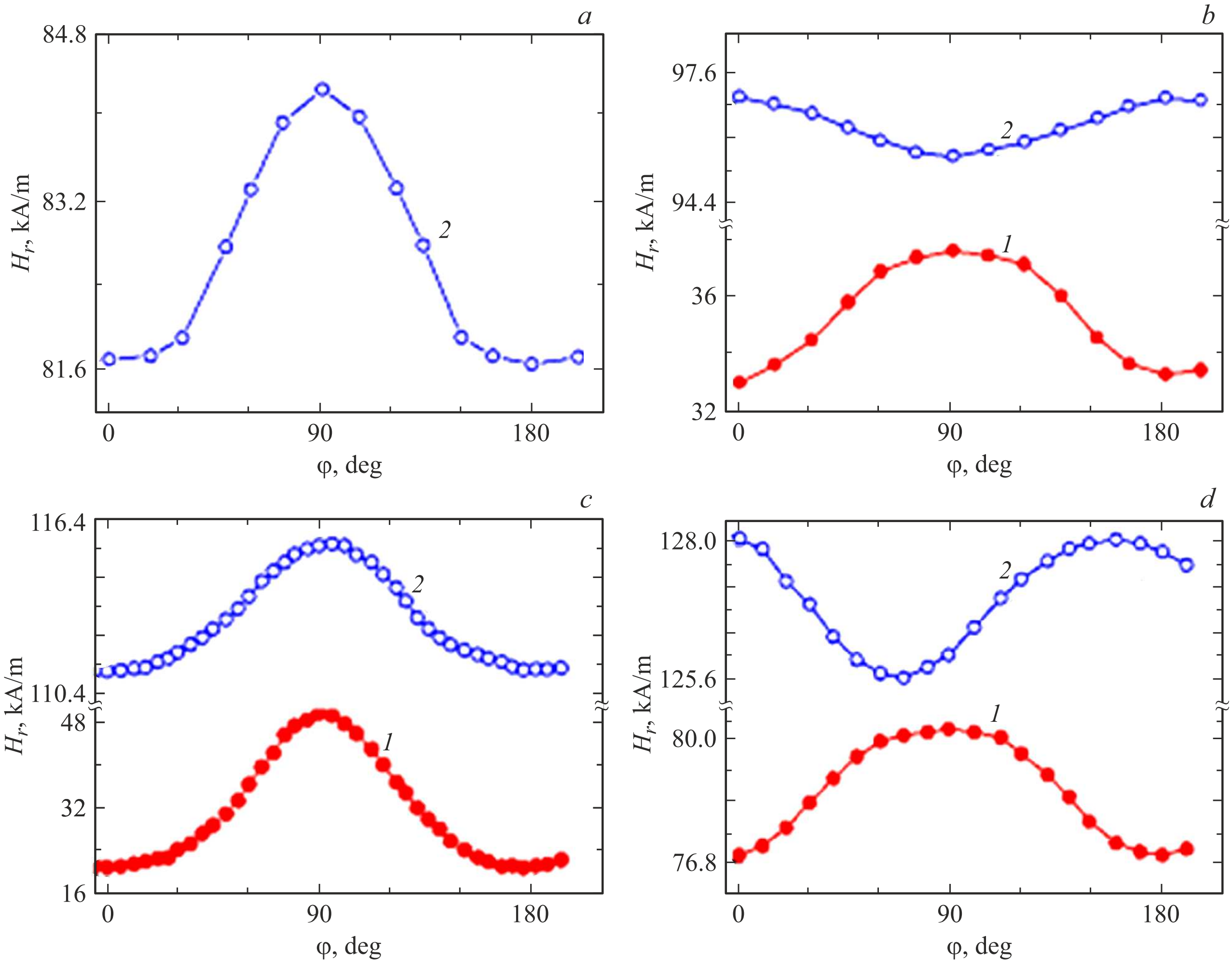


**Figure 3.** Angular dependences of the magnitudes of the resonance fields of CoNi/Si/FeNi films. $t_{\mathrm{Si}} = 2.4$ $(a, c)$ and 3.3 nm $(b, d)$. $T = 10$ $(a, b)$ and 293 K $(c, d)$. *1, 2* — peaks assigned to CoNi and FeNi layers, respectively. The magnetic field is in the film plane.

100 A/m, so the magnetization orientation of this layer in the three-layer structure will be determined by the magnetic interactions with the CoNi layer and the external magnetic field. The magnetization orientation of the CoNi layer is set by the sub-magnetizing field during the sputtering process, which determines the light magnetization anisotropy axis. In the absence of an external magnetic field, the magnetically hard layer (CoNi) is oriented along the light direction, and the direction angle of the soft magnetic layer (FeNi) will be determined by the ratio $\cos(\varphi_{\mathrm{FeNi}}) = -J_1/(2J_2)$ [15], where $J_1$ and $J_2$ — constants of bilinear and biquadratic interlayer interactions (assuming negative biquadratic interaction). The position of the maximum of the resonance field of the line *2* on the angular dependence at $T = 10$ K falls at $\varphi_{\mathrm{FeNi}} = \pi/2$, so, $|J_2| \gg |J_1|$. The fact that this maximum shifts with temperature change means that the interlayer interaction parameters are temperature-dependent.

Currently, the main mechanisms responsible for the occurrence of the biquadratic interaction are: 1) quantum interference [16]; 2) fluctuation mechanism [17]; 3) „loose spin“ model [18]; 4) magnetodipole mechanism [19]. If we take into account the ratio of the $J_1/(2J_2)$ values in our case, the mechanisms 1 and 2 lead to an oscillatory nature of the interaction depending on the nonmagnetic layer thickness, but the contribution of the biquadratic interaction is insufficient for the observed effect. Mechanism 4 results in an oscillation of the interaction in the lateral direction and a decreasing magnitude as a function of the nonmagnetic layer thickness. The theory based on loose interface spins [18] (mechanism 3) basically describes the dependence on both the nonmagnetic layer thickness and temperature. However, in our case (nonmagnetic semiconductor layer), the latter circumstance is most likely related to the semiconductor properties, namely, to the increase in the number of charge carriers in the nonmagnetic layer and, as a consequence, the number of interaction carriers between magnetic layers.

Thus, in this paper, it is found that the presence of a silicon semiconductor interlayer as part of a three-layer structure with magnetically hard (CoNi) and soft magnetic (FeNi) layers has a significant effect on the magnetic state of the structure, namely:

— the thickness of the semiconductor interlayer affects the interlayer interaction in the CoNi/Si/FeNi trilayer structure;

— the biquadratic interlayer interaction between the magnetically hard and soft magnetic layers is found to occur between the magnetically hard and soft magnetic layers by direct measurements;

— the parameters of the interlayer interaction are temperature dependent.

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### Conflict of interest

The authors declare that they have no conflict of interest.

## References

[1] J.A.C. Bland, A. Hirohata, C.M. Guertler, Y.B. Xu, M. Tselepi, J. Appl. Phys., **89** (11), 6740 (2001). DOI: 10.1063/1.1361045
[2] P. Grünberg, D.E. Bürgler, H. Dassow, A.D. Rata, C.M. Schneider, Acta Mater., **55** (4), 1171 (2007). DOI: 10.1016/j.actamat.2006.10.002
[3] B. Heinrich, in *Magnetic heterostructures. Advances and perspectives in spinstructures and spintransport*, ed. by H. Zabel, S.D. Bader (Springer, Berlin–Heidelberg, 2008), p. 185. DOI: 10.1007/978-3-540-73462-8_4
[4] M. Ruhrig, R. Schafer, A. Hubert, R. Mosler, J.A. Wolf, S. Demokritov, P. Grunberg, Phys. Status Solidi A, **125** (2), 635 (1991). DOI: 10.1002/pssa.2211250225
[5] U. Köbler, K. Wagner, R. Wiechers, A. Fuss, W. Zinn, J. Magn. Magn. Mater., **103** (3), 236 (1992). DOI: 10.1016/0304-8853(92)90195-T
[6] A.B. Drovosekov, O.V. Zhotikova, N.M. Kreines, D.I. Kholin, V.F. Meshcheryakov, M.A. Milyaev, L.N. Romashev, V.V. Ustinov, JETP, **89** (5), 986 (1999). DOI: 10.1134/1.558941.
[7] A.B. Rinkevich, M.A. Milyaev, L.N. Romashev, Phys. Met. Metallogr., **120** (3), 247 (2019). DOI: 10.1134/S0031918X19030116.
[8] S.O. Demokritov, A.B. Drovosekov, D.I. Kholin, N.M. Kreines, J. Magn. Magn. Mater., **258-259**, 391 (2003). DOI: 10.1016/S0304-8853(02)01123-X
[9] S.O. Demokritov, A.B. Drovosekov, D.I. Kholin, N.M. Kreines, H. Nembach, M. Rickart, J. Magn. Magn. Mater., **272-276**, E963 (2004). DOI: 10.1016/j.jmmm.2003.12.1339
[10] C.J. Gutierrez, J.J. Krebs, M.E. Filipkowski, G.A. Prinz, J. Magn. Magn. Mater., **116** (3), L305 (1992). DOI: 10.1016/0304-8853(92)90106-X
[11] E.E. Fullerton, S.D. Bader, Phys. Rev. B, **53** (9), 5112 (1996). DOI: 10.1103/PhysRevB.53.5112
[12] G.J. Strijkers, J.T. Kohlhepp, H.J.M. Swagten, W.J.M. de Jonge, Phys. Rev. Lett., **84** (8), 1812 (2000). DOI: 10.1103/PhysRevLett.84.1812
[13] G.S. Patrin, I.A. Turpanov, V.I. Yushkov, A.V. Kobyakov, K.G. Patrin, G.Yu. Yurkin, Ya.A. Zhivaya, JETP Lett., **109** (5), 320 (2019). DOI: 10.1134/S0021364019050126.
[14] S. Tumanski, *Handbook of magnetic measurements* (CRC Press, Boca Raton, 2011). DOI: 10.1201/b10979
[15] S.O. Demokritov, J. Phys. D: Appl. Phys., **31** (8), 925 (1998). DOI: 10.1088/0022-3727/31/8/003
[16] P. Bruno, Phys. Rev. B, **52** (1), 411 (1995). DOI: 10.1103/PhysRevB.52.411
[17] J.C. Slonczewski, Phys. Rev. Lett., **67** (22), 3172 (1991). DOI: 10.1103/PhysRevLett.67.3172
[18] J.C. Slonczewski, J. Appl. Phys., **73** (10), 5957 (1993). DOI: 10.1063/1.353483
[19] S.O. Demokritov, E. Tsymbal, P. Grunberg, W. Zinn, I.K. Schuller, Phys. Rev. B, **49** (1), 720 (1994). DOI: 10.1103/PhysRevB.49.720

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