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ORIGINAL PAPER

Influence of the Dy_xCo_{1-x}–Bi Interface on the Magnetic Properties of Dy_xCo_{1-x}/Bi/Py Three-Layer Structures

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Abstract
The interlayer interactions in multilayer systems with a non-magnetic semimetallic interlayer are great of interest. The magnetic and structural properties of the Dy_xCo_{1-x}/Bi/Py systems ($17 < x < 26$ at.%) have been studied. The temperature dependences of magnetization in the range 4.2–300 K were measured for the first time. The influence of the bismuth interlayer thickness on the exchange interaction between the DyCo and Py layers was found as well as the critical value of its thickness. The obtained atypical value of the period of exchange bias oscillations was explained by the formation of bismuth compounds with dysprosium–pnictogenides at bismuth thicknesses below the critical value. The interface was investigated by spectral ellipsometry in the range 2–5 eV. The information on the structure of the surface obtained by atomic force microscopy was used to create a multilayer model for fitting experimental ellipsometric data. Analysis of the optical properties showed that pnictogenide Dy_3Bi_2 is formed at the interface, which affects the general magnetic state of the samples studied.

Keywords Multilayer magnetic films · Exchange interaction · Exchange bias · Ellipsometry · SQUID magnetometry

1 Introduction

Amorphous alloys of rare earth metals with 3d transition metals are currently of significant research interest due to the variety of interesting physical properties and their possible practical applications. Such alloys can be used to make storage media with a high recording density, new types of magnetoresistive memory, and high sensitive magnetic field sensors [1]. The unusual properties of rare earth elements (e.g., large values of the magnetic moment and coercive force) can be used to design new materials based on dilute magnetic semiconductors [2].

At present, interlayer interactions in multilayer systems with a non-magnetic semimetallic interlayer are poorly studied. The work with such systems is relevant when studying the spin valve structures and giant magnetoresistance effect [3]. Bismuth is a promising material for use as a non-magnetic interlayer because it has a number of unusual physical properties. This semimetal, according to the phase diagram [4], does not form compounds with most 3d metals. Thus, the absence of bismuth-3d-metal compounds is expected in the films [5]. In addition, bismuth is characterized by a high anisotropy of its Fermi surface, a low concentration of charge carriers, and a low effective mass [6]. Its Fermi wavelength is 40 nm [7]. Bismuth is also characterized by a large electron mean free path, and as a result, it has been studied for finite size effects and for quantum transport.

Several works were devoted to research some aspects of the exchange interaction in three-layer systems with a bismuth interlayer [7–10]. Anisotropic behavior and an inflection of the magnetization curve found in Fe/Bi/Fe three-layer systems were explained by interlayer interaction existing through the bismuth interlayer [8]. Oscillations of the exchange interaction depending on bismuth thickness with period changing from 180 to 200 Å were observed in CoFe/Bi/Co films at room temperature [9]. The authors also noted

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that with the temperature decreasing the interlayer interaction increased oscillating with the Bi thickness in a similar way, but the oscillation amplitude decreased.

In addition to the influence of the bismuth layer thickness on the interlayer interaction, the dependence of the saturation magnetization on the bismuth layer thickness was also found in three-layer NiFe/Bi/NiFe films [10], which can be associated with the effect of spin accumulation in the semimetal.

Varying the thickness of the bismuth interlayer did not affect the magnetic and magneto-optical properties of Co(5 nm)/Bi/Co(5 nm) films [7]. In this case, the coercive force and saturation field of the films increased with the thickness of the bismuth layer increasing.

However, to date, there is no complete understanding of the exchange interaction features in thin-film magnetic systems with a semimetallic Bi layer.

Previously, we synthesized three-layer systems Dy_xCo_{1-x}/Bi/Py [11], consisting of layers of ferromagnetic permalloy (Py) and ferrimagnetic alloy Dy_xCo_{1-x} separated by a non-magnetic semimetallic bismuth interlayer. Their magnetic properties were studied using the magneto-optical Kerr effect. We found that the bismuth interlayer thickness is responsible for the temperature of the magnetic compensation point in the Dy_xCo_{1-x} layer. It was attributed to the influence of the interface precisely at the Dy_xCo_{1-x}–Bi interface, since as shown earlier [12], the bismuth–permalloy interface weakly affects the magnetic properties of the structure. However, the possible presence of secondary phases at the Dy_xCo_{1-x}–Bi interface remained unclear, since even the very presence of a pure DyCo/Bi interface can lead to a significant change in the magnetic state in the DyCo film due to the strong spin–orbit interaction of bismuth.

In contrast to our previous work [11], the present study uses the integral magnetization measured from the entire volume of the Dy_xCo_{1-x}/Bi/Py three-layer structure.

2 Materials and Methods

The synthesis of the Dy_xCo_{1-x}/Bi/Py films was carried out by thermal evaporation in vacuum (the base vacuum was $2 \cdot 10^{-6}$ mbar) and included successive deposition of Py-layer ($Ni_{1-x}Fe_x$, $18 < x < 23$ at.%), bismuth layer, and the alloy Dy_xCo_{1-x} ($17 < x < 26$ at.%). The temperature dependences of the magnetization were measured using an MPMS XL (Quantum Design) magnetometer working in the temperature range $T = 4.2$–300 K. The structure of the Dy_xCo_{1-x}–Bi interface was analyzed using Veeco MultiMode NanoScope IIIa SPM System atomic force microscope measurements, as well as the spectra of ellipsometric parameters Ψ and Δ measured on the Spectroscan ellipsometer in the spectral range 2–5 eV.

3 Results and Discussion

Figure 1 shows the results of measuring the temperature dependences of the saturation magnetization. It shows a monotonic character (except for $T < 20$ K, where there is a sharp decrease in the magnetization for all thicknesses) in the absence of a bismuth interlayer, as well as for the thicknesses 1 and 5 nm. However, for bismuth thickness $d_{Bi} = 2$ nm, the magnetization increases starting from the temperature ~ 100 K and has a fairly extended maximum in the region 150–180 K, which corresponds to the compensation temperature in the DyCo layer, which we measured in previous work [11]. It should be noted that the magnetization values in this area for the film with $d_{Bi} = 2$ nm coincide with the magnetization for the film with $d_{Bi} = 5$ nm. As can be seen from Fig. 1, at a fixed temperature, oscillations of the magnetization value are observed depending on the thickness of the non-magnetic layer. Against this background, the behavior of the film with $d_{Bi} = 2$ nm stands out somewhat, which may be due to the anisotropy of the magnetic subsystems, where the DyCo subsystem plays the main role in the formation of anisotropic properties. The temperature variation of magnetizations for films with $d_{Bi} > 3$ nm is monotonic in the range of 85–300 K and is apparently associated with a weakening of the interlayer interactions.

The field dependences of the magnetizations in Fig. 2 show that as the interlayer increases (see parts c and d in Fig. 2), the hysteresis loops undergo smaller changes with an increasing temperature. It is known that the resulting magnetization M of DyCo layer is composed of antiparallel magnetizations of the rare earth metal (Dy) and the transition metal (Co) sublattices. In the temperature range 150–180 K, magnetization compensation point arises, which affects the total magnetization of the three-layer structure.

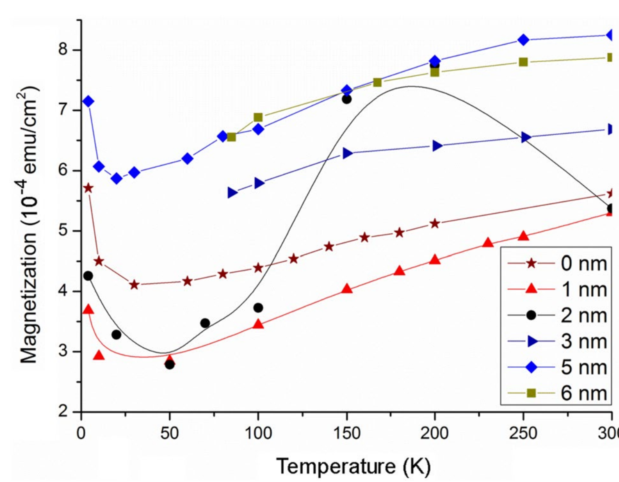

Fig. 1 Temperature dependences of saturation magnetization in systems Dy_xCo_{1-x}/Bi/Py for different thicknesses of the bismuth interlayer

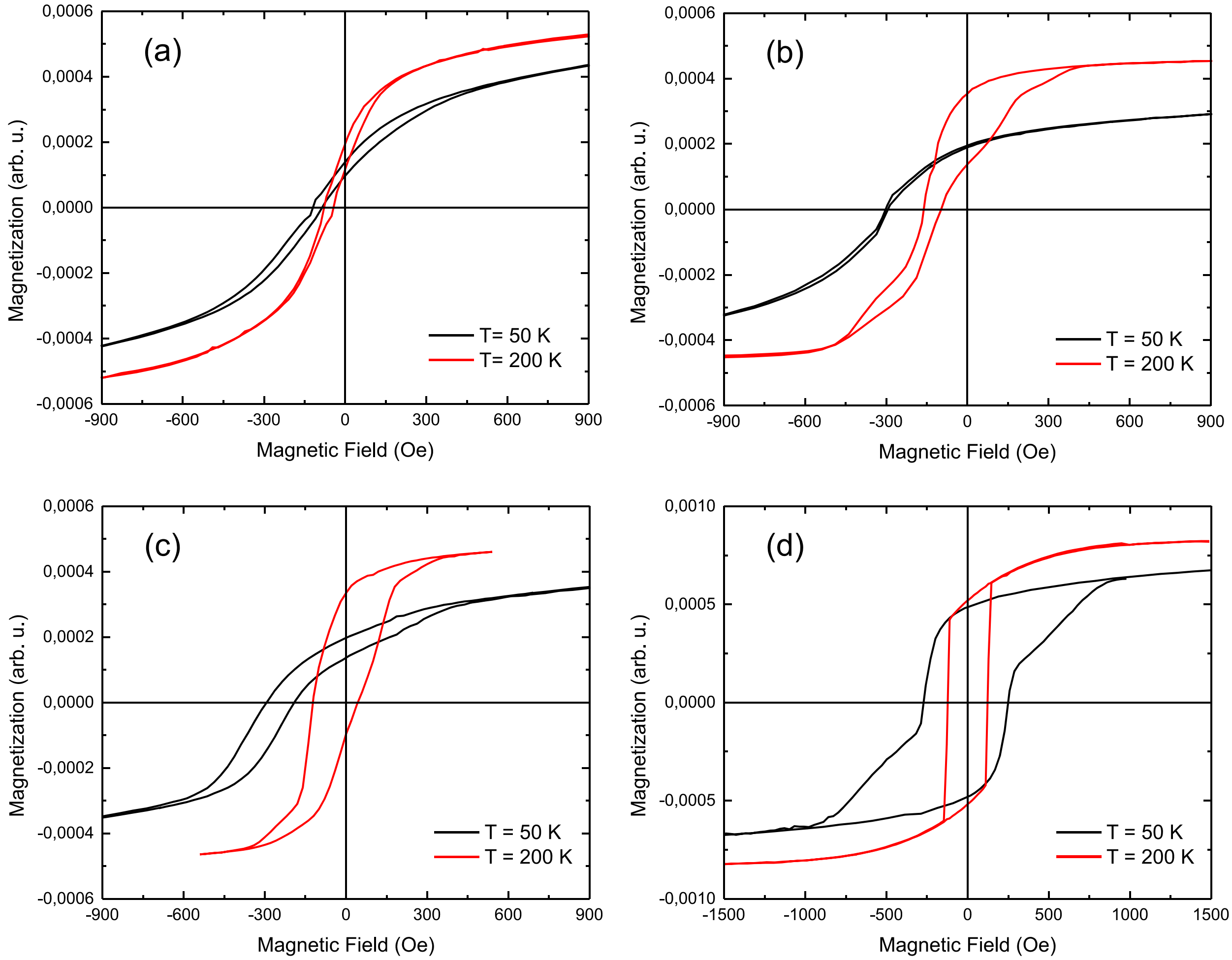

Fig. 2 Magnetization curves at temperatures of 50 and 200 K for different thicknesses of the bismuth interlayer: **a** $d_{Bi}=0$ nm, **b** $d_{Bi}=1$ nm, **c** $d_{Bi}=2$ nm, **d** $d_{Bi}=5$ nm

For the systems studied, temperature 50 K is obviously less than any value of the compensation temperature, and temperature 200 K is the maximum value of the compensation temperature corresponding to a single-layer DyCo film [11]. Starting from the critical interlayer thickness $d_{Bi}=2$ nm, the coercive force increases at low temperatures, while at the bismuth thickness $d_{Bi}=1$ nm, the coercive force is lower than in the sample without an interlayer. Moreover, the thickness of the interlayer affects the exchange bias field. The nature of the exchange bias effect in the DyCo film can be associated with different magnetic behavior on the surface and inside the layer. It was shown [13] that the effect of Co on the magnetic properties of the sample dominates in the bulk of the film, while Dy has a significant effect on the magnetic properties on the film surface due to a strong decrease in the magnetization of Co on the surface. On the other hand, it is known that in film systems with bismuth, as the layer thickness decreases, a transition from a semimetallic to a semiconductor state is possible, which can affect the magnitude of interlayer interactions [14]. However, this issue requires special research, which will be done on the basis of measurements of magnetically dependent transport.

In the temperature region of the maximum magnetization, the coercive force H_c starts to increase dramatically for the thickness of the interlayer $d_{Bi}=2$ nm (Fig. 3). At room temperature, the H_c values for all samples (including the two-layer sample without a bismuth interlayer) differ by no more than 30 Oe. Therefore, the effect of the interlayer on the overall magnetic state is minimal at $T=300$ K.

Next, it is worth noting the contradiction that the magnetization value M for $d_{Bi}=0$ nm lies between M for $d_{Bi}=1$ nm and M for $d_{Bi}=5$ nm, although exchange interaction oscillations are expected at large thicknesses [8]. We may assume the materials that make up the interlayer between the ferri- and ferromagnetic layers are different for each sample. So at a small thickness, layers all or almost all the bismuth interacts with neighboring. Given that the interaction of Bi with 3d metals can be excluded, it is worth considering

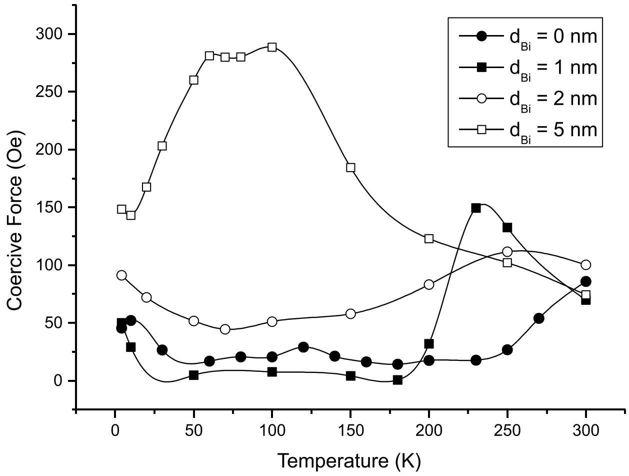

Fig. 3 Temperature dependence of the coercive force for different thicknesses of bismuth

compounds of bismuth with dysprosium, i.e., pnictogenides (Dy_5Bi_3, Di_3Bi_2, or DyBi).

To accurately determine the type of pnictogenide, ellipsometric measurements were carried out. To approximate the ellipsometric data in the first approximation, a multilayer model [14] was used, in which the Fresnel coefficients of each subsequent layer are related to the coefficients of the previous one by the following recursive relation:

$$r_{j+1} = \frac{r_{j+1}(1 - r_j r_{j+1}) + (r_j - r_{j+1})X}{1 - r_j r_{j+1} + r_{j+1}(r_j - r_{j+1})X}, \quad (1)$$

where $X = \exp(-2i\delta)$, δ is the phase thickness of the corresponding layer [15], and r_j is the Fresnel coefficients of the corresponding interfaces. It should be noted that the equation is valid for both *p* and *s* polarizations. However, a significant drawback of this model is the presence of sharp interfaces between the layers, which most often do not correspond to reality. The pnictogenides formed in Bi–Py interfaces must be taken into account in any case when fitting ellipsometric data. For this purpose, the equivalent film method [15] was used as follows. An additional layer is introduced into the multilayer model, which approximates the connection with a mixture of two materials. If the volume fractions of Dy and Bi are known, then the effective complex refractive index N_{eff} of such a layer can be calculated numerically from the equation:

$$q\frac{N_f^2 - N_{eff}^2}{N_f^2 + 2N_{eff}^2} + (1-q)\frac{N_a^2 - N_{eff}^2}{N_a^2 + 2N_{eff}^2} = 0, \quad (2)$$

where N_f and N_a are the refractive indices of the two materials, and q is the relative volume occupied by the materials.

Table 1 Average values of the roughness parameters of the DyCo surface

RMS (nm)	0.784
Max height Rmax (nm)	2.061
Rz (nm)	2.046

In order to build a model of the interface layer, the surface of the samples was examined by atomic force microscopy (Fig. 4). Table 1 shows the roughness parameters (the standard deviation of the roughness profile (RMS) and the height of irregularities determined by 10 main points (Rz) of the surface. It should be noted that the granule size is quite large (70–80 nm) along the film plane. While the size of the roughness coincides with the characteristic thickness of the bismuth interlayer, on which critical changes in the magnetic properties occur. These parameters were used to fit the ellipsometric spectra and made it possible to get a good agreement between the theoretical and experimental curves. As a result, the inverse problem of ellipsometry was solved for the measured spectra of the ellipsometric parameters Ψ and Δ. Figure 5 shows the spectral dependence of Ψ, Δ, and the results of modeling by the Brugeman method [15]. The solution of the inverse problem for all samples showed that

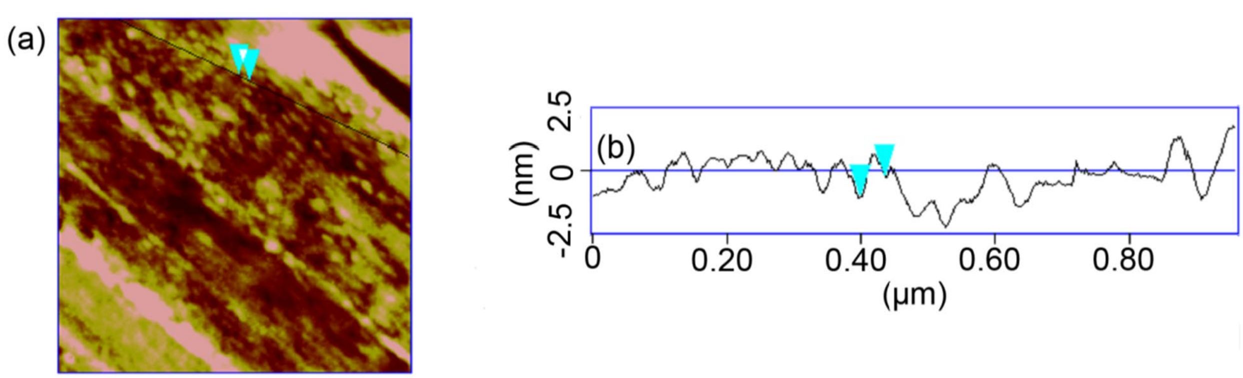

Fig. 4 Results of studying the surface of the DyCo layer by atomic force microscopy: **a** AFM topography image, **b** profile image demonstrating surface roughness

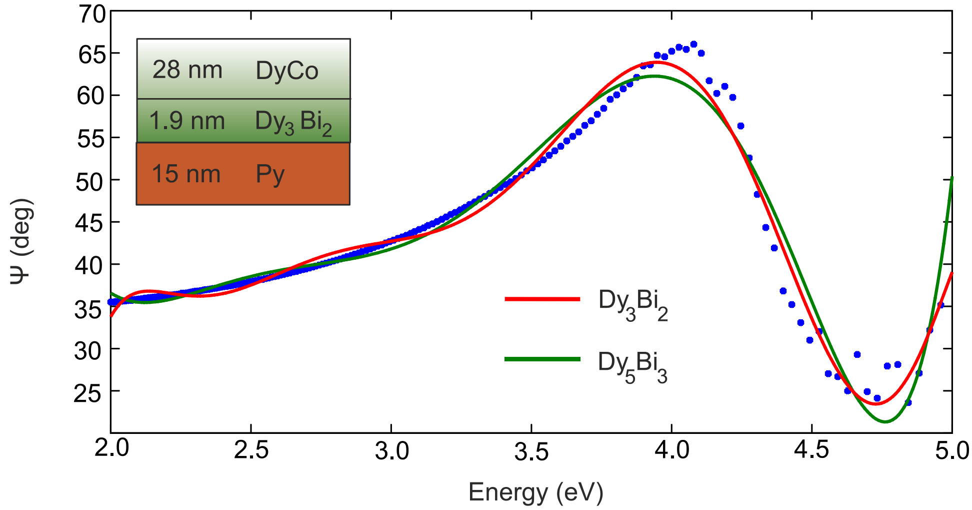

Fig. 5 Spectrum of the ellipsometric parameter Ψ for a sample with an interlayer $d_{Bi} = 2$ nm; the insert shows scheme of a three-layer system

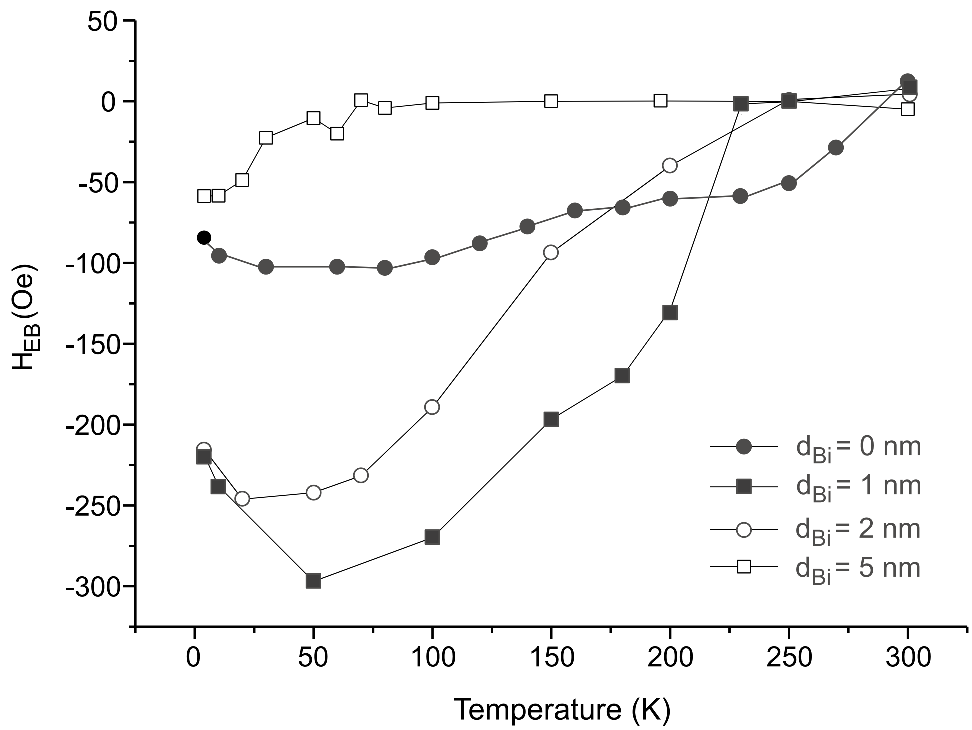

Fig. 6 Temperature dependence of the exchange bias field for different thicknesses of the bismuth interlayer

at interlayer thicknesses < 2 nm, this layer is a mixture of bismuth and dysprosium in a ratio of 2:3.

As the bismuth thickness increases to 5 nm, the "magnetization" of the hard magnetic layer by the soft magnetic layer weakens, and, consequently, the interlayer thickness becomes too large for the exchange between DyCo and Py to affect the magnetization processes in DyCo. This is confirmed by the temperature dependence of the exchange bias field H_{EB} (Fig. 6), which is close to zero for thickness $d_{Bi} = 5$ nm at $T > 50$ K. For smaller interlayer thicknesses, we observed a nonlinear behavior of the exchange bias with thickness, although exchange interaction oscillations are not expected here, which also confirms the formation of a complex interface.

4 Conclusion

Thus, analyzing of obtained temperature dependences of the saturation magnetization for different interlayer thicknesses allows to assume that the bismuth thickness 2 nm is critical when the influence of the interlayer on the exchange interaction becomes decisive.

The presence of secondary phases at the Dy_xCo_{1-x}–Bi interface with the formation of bismuth-dysprosium compounds called pnictogenides at bismuth thicknesses below the critical value leads to an atypical value of the period of exchange bias oscillations.

The analysis of Dy_xCo_{1-x}–Bi interface carried out by atomic force microscope measurements and spectral ellipsometry showed that the pnictogenide Dy_3Bi_2 is formed at the interface, which affects the general magnetic state of Dy_xCo_{1-x}/Bi/Py.

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Data Availability The data that support the findings of this study are available from the corresponding author, Nikolai Kosyrev, upon reasonable request.

Declarations

Competing Interests The authors declare no competing interests.

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