## 01,05,13

# Spin-flop transition near the temperature of magnetic compensation in amorphous ferrimagnetic Dy-Co films obtained by ion-plasma sputtering

© A.S. Rusalina<sup>1</sup>, V.N. Lepalovskij<sup>1</sup>, E.A. Stepanova<sup>1</sup>, A.A. Yushkov<sup>1</sup>, A.N. Gorkovenko<sup>1</sup>, E.V. Kudyukov<sup>1</sup>, V.O. Vas'kovskiy<sup>1,2</sup>, G.V. Kurlyandskaya<sup>1</sup>, A.V. Svalov<sup>1</sup>

 <sup>1</sup> Institute of Natural Sciences and Mathematics, Ural Federal University named after the First President of Russia B.N. Yeltsin, Yekaterinburg, Russia
 <sup>2</sup> M.N. Mikheev Institute of Metal Physics, Ural Branch, Russian Academy of Sciences, Yekaterinburg, Russia

E-mail: anastasia.rusalina@urfu.ru, andrey.svalov@urfu.ru

Received April 18, 2024 Revised April 18, 2024 Accepted May 8, 2024

A spin-flop transition was observed in amorphous ferrimagnetic Dy-Co, films obtained by ion-plasma sputtering near the temperature of magnetic compensation. The temperature dependence of the critical field is described in the framework of the molecular field theory for a two-lattice ferrimagnet. An estimate of the effective constant of the inter-lattice exchange interaction is made.

Keywords: amorphous magnetic films, ferrimagnetism, phase transition, magnetic anisotropy.

DOI: 10.61011/PSS.2024.06.58687.10HH

## 1. Introduction

The amorphous ferrimagnetic films of alloys of heavy rare earth elements (RE) and transition 3d-metals (TM) have again drawn an increased attention of researchers in recent years. This, in particular, is attributable to the demonstrated possibility of changing the orientation of the magnetization of the RE-TM film using ultrashort optical pulses without applying an external magnetic field [1]. It has recently been shown that the features of this remagnetization process depend on the rare earth component of the film, including its magnetic anisotropy [2]. In addition, the phase transition from a collinear to a non-collinear spin state can significantly change the ultrafast spin dynamics in ferrimagnetic films RE-TM [3].

Dy-Co films are characterized by nonmonotonic concentration dependences of the main magnetic properties. These materials have a characteristic sperimagnetic structure with a Co content of more than 50,at.%, and a quantitative description of spontaneous magnetization is possible in the molecular field model jcite4. A magnetic fieldinduced spin-flop transition can be observed in Dy-Co films near the magnetic compensation temperature  $T_{\rm comp}$  [5]. This paper is devoted to discussing the features of the spin-flop transition in amorphous ferrimagnetic films of Dy-Co obtained by ion-plasma sputtering. To do this, the magnetic properties of the films were studied in a wide temperature range, including the temperature of magnetic compensation.

# 2. Study methodology

Dy18.7Co81.3 films were obtained by co-sputtering on Corning glass substrates with simultaneous ion-plasma sputtering of Dy and Co targets in an argon atmosphere. The composition of the films was controlled by regulating the power supplied to the targets. The deposition was carried out on a rotating substrate at a rotation speed of 40 rpm for increasing the uniformity of the composition over the area of the films. The thickness of the films was 90 nm at a deposition rate of 9 nm/min. All samples were protected from oxidation by 5 nm thick buffer and coating Ta layers. The films were deposited in the presence of a constant magnetic field of 250 Oe, oriented in the substrate plane. Transmission electron microscopy (TEM) with JEM-2100 microscope was used for structural studies of the films at an accelerating voltage of 200 kV. The samples intended for structural studies were deposited on fresh chips of NaCl single crystals. The magnetic properties were studied using the measuring system MPMS-7XL in the temperature range from 5K to 300K and the magneto-optical Evico Kerr microscope at a room temperature.

# 3. Results and discussion

The results of studies using transmission electron microscopy demonstrate an amorphous structure of the samples. Figure 1, *a* shows a TEM image of  $Dy_{18.7}Co_{81.3}$  film. It shows a weak contrast characteristic of an amorphous structure and the absence of microcrystallites. The blurred



**Figure 1.** TEM image (a) and microdiffraction pattern (b) of the film  $Dy_{18.7}Co_{81.3}$ .

halos observed in the diffraction pattern of the selected area (Figure 1, b) confirm this conclusion.

Hysteresis loops measured along and perpendicular to the sample plane indicate that the films have effective planar magnetic anisotropy. Figure 2 shows as an example some hysteresis loops of a film of  $Dy_{18.7}Co_{81.3}$ ,measured using a SQUID magnetometer at different temperatures and orientations of the external magnetic field parallel to the sample plane. In turn, an induced magnetic anisotropy was detected in the plane of the samples (Figure 3), the easy axis of which was parallel to the axis of the magnetic field present during film deposition. The mechanism of occurrence of this so-called M-induced anisotropy is well known [6,7]. The value of the anisotropy field for this film was approximately 2000 Oe, which corresponds to the anisotropy constant of  $K \approx 1.8 \cdot 10^5$  erg/cm<sup>3</sup> at M = 180 G.

The temperature dependence of the spontaneous magnetization M(T) was determined based on measurements of magnetometric hysteresis loops at different temperatures. It was assumed that the value of the residual magnetization corresponds to the value of spontaneous magnetization. A low characteristic of ferrimagnets was observed on this dependence near the compensation temperature (Figure 4). The ferrimagnetic nature of the studied films is also confirmed by the characteristic temperature dependence of the coercive force  $H_c(T)$  with a local maximum near  $T = T_{comp}$ (Figure 5). The analysis of the dependencies M(T) and  $H_c(T)$  allows concluding that for the film  $Dy_{18.7}Co_{81.3}$  $T_{comp} \approx 175$  K.

Fractures were observed at temperatures near  $T_{\text{comp}}$  on magnetometric hysteresis loops at a certain magnitude of the external magnetic field  $H = H_{\text{cr}}$  (Figure 2), which is characteristic of the spin-flop transition and the occurrence of a non-collinear magnetic structure in ferrimagnets [8]. The data obtained for  $H_{\text{cr}}$  are summarized in Figure 6.

It is shown that the value of  $H_{\rm cr}$  depends on temperature and decreases as it approaches  $T_{\rm comp}$ . The expression for  $H_{\rm cr}$ is written as follows for a two-lattice ferrimagnet RE-TM in the framework of molecular field theory [8]:

$$H_{\rm cr} = \lambda |M_{\rm RE} - M_{\rm TM}|, \qquad (1)$$

where  $M_{\rm RE}$  and  $M_{\rm TM}$  — the magnetization of the sublattices of rare earth and transition metals, respectively;  $\lambda$  — the constant of the interlattice exchange interaction. Solid lines in Figure 6 are constructed based on experimental data using the least squares method. The lines intersect at  $T \approx 175 \text{ K} = T_{\text{comp}}$ . In addition, the rate of change of  $H_{\rm cr}$  with temperature is the same at  $T < T_{\rm comp}$  and  $T > T_{\text{comp}}$  ( $|\Delta H_{\text{cr}}|/\Delta T = 0.6 \text{ kOe/K}$ ), and this behavior corresponds to the same rate of change of  $M_s$  near  $T_{\rm comp}$  $(|\delta M_s|/\Delta T = 1.6 \,\text{G/K})$ . This can be considered as additional evidence that the magnetic properties of amorphous ferrimagnetic films of Dy-Co can be satisfactorily described within the framework of the molecular field theory for a two-lattice ferrimagnet. It should be noted that it follows from the equation (1) that the value of  $H_{cr}$  decreases as it approaches the temperature of magnetic compensation and turns to zero at  $T = T_{\text{comp}}$  when  $|M_{\text{RE}} - M_{TM}| = 0$ . The experimentally observed value  $H_{cr}$  is minimal near  $T_{comp}$ , but is different from zero (Figure 6). Most likely, this is attributable to the fact that the expression (1) is true for an isotropic ferrimagnet, while the induced magnetic anisotropy was formed in the films studied in this paper in the plane of the films as a result of sputtering in the presence of a constant magnetic field. Moreover, the impact of possible chemical heterogeneity in the volume of the sample is also possible.

The values of the constant of the intersublattice interaction were calculated according to the following expression based on the sections of hysteresis loops in the region of fields exceeding  $H_{\rm cr}$  [5,8]:

$$\lambda = \partial H / \partial M. \tag{2}$$

The averaging of the results obtained from several loops measured at different temperatures yields the value of  $\lambda \approx 1000$ .



**Figure 2.** Hysteresis loops of  $Dy_{18.7}Co_{81.3}$  film measured at different temperatures and orientations of the magnetic field in the sample plane.

There is another way to estimate  $\lambda$ . It is known that the dependence M(T) can be adequately described within the framework of the phenomenological theory of spontaneous magnetization for amorphous ferrimagnetic films based on the molecular field model [4,9–12] in the following form

$$M_{s}(x, T) = |M_{\text{RE}}(x, T=0)B_{R}(\xi_{R}) - M_{\text{TM}}(x, T=0)B_{T}(\xi_{T})|,$$
(3)

where Mi(x, T = 0) — sublattice magnetizations of the ground state,  $B_i(\xi_i)$  — Brillouin functions describing the temperature change of sublattice magnetizations, x — concentration of the rare earth component. The molecular field for each magnetic sublattice has an intra- and inter-

sublattice components and is determined by the following expression

$$H_i^* = \lambda_{ii}(x)M_i(x,T) + \lambda_{ij}(x)M_j(x,T), \qquad (4)$$

where  $\lambda_{ii}(x)$  and  $\lambda_{ij}(x)$  are constants of intra- and intersublattice interaction, which, in turn, are reduced to exchange integrals  $J_{\text{TM-TM}}$ ,  $J_{\text{TM-RE}}$ ,  $J_{\text{RE-RE}}$  [10]. These values are parameters of the phenomenological theory and are determined by approximating the experimental dependencies  $M_s(T)$  using the formula (3). The relationship between the constant of the inter-lattice interaction  $\lambda = \lambda_{ij}$ 



**Figure 3.** Magneto-optical hysteresis loops of  $Dy_{18.7}Co_{81.3}$  film measured in the sample plane along (*I*) and perpendicular to the easy axis (2). The insert shows a domain structure formed in a field close to the coercive force of the sample.



**Figure 4.** Experimental (points) and calculated (line) temperature dependence of spontaneous magnetization for the  $Dy_{18.7}Co_{81.3}$  film.

and  $J_{\text{TM-RE}}$  is determined by the expression [5,8]:

$$\lambda_{ij} = \frac{2J_{ij}\langle Z \rangle}{Ng^2 \mu^2},\tag{5}$$

where Z — the number of nearest neighbors of atoms; N — the number of atoms per unit volume;  $g_{RE}$  and  $g_{TM}$  — gyromagnetic factor for RE and TM elements, respectively;  $\mu_B$  — Boron magneton. A solid line in Figure 4 shows the calculated temperature dependence of the spontaneous magnetization for the film of Dy<sub>18.7</sub>Co<sub>81.3</sub>. A good agreement between the calculated and experimental data was obtained at the value of the fitting parameter  $J_{TM-RE} = 23 \cdot 10^{-16}$  erg. The sperimagnetic structure of the film of Dy-Co was taken into account in the calculation according to the procedure described in Ref. [7]. The order of the obtained parameter  $J_{\text{TM-RE}}$  agrees well with the data for other amorphous films of RE-TM [4,9–12].  $\lambda \approx 1000$  is found according to the expression (5) using this value  $J_{\text{TM-RE}}$  and assuming that  $g_{\text{RE}} = 4/3$ ,  $g_{\text{TM}} = 2$ . Thus, two different ways of determining  $\lambda$  give the same result.

The obtained values  $\lambda \cong 1000$  coincide with the value found for the single crystal DyCo<sub>5.3</sub> [8]. At the same time, they are approximately five times higher than the values  $\lambda$ obtained in Ref. [5] for a similar composition of the film of Dy-Co prepared by thermal sputtering. Such a difference in the values of  $\lambda$  is most likely due to the microstructure of films obtained by different methods, namely, the features of the short-range order, and above all, variations in the interatomic distance. In addition, it was suggested that the microstructure of Dy-Co films obtained by thermal sputtering may constitute a set of nanoscale crystallites of



**Figure 5.** The temperature dependence of the coercive force of the film  $Dy_{18.7}Co_{81.3}$ .



**Figure 6.** The temperature dependence of the critical field for  $Dy_{18.7}Co_{81.3}$  film obtained when the magnetic field is oriented in the plane of the sample.

Dy<sub>x</sub>Co<sub>y</sub> of the corresponding compositions, wherein the size of the crystallites is units of nanometers, which is characterized as an X-ray amorphous state [14]. In this case, it is possible that the fractures on the magnetization curves for thermal films of Dy-Co are not a reflection of the spin-flop transition, but of a violation of the magnetic structure of the sample due to the exchange interaction between crystallites of different composition, similar to how it was observed in inhomogeneous films of Gd-Co [15]. It is logical to assume that the intensity of the exchange interaction between the crystallites is less than the intensity of the exchange interaction between RE and TM sublattices, which results in a lower value of the effective  $\lambda$ .

## 4. Conclusion

A spin-flop transition was recorded in amorphous ferrimagnetic films of Dy-Co obtained by ion-plasma sputtering near the temperature of magnetic compensation, characteristic of a two-sublattice ferrimagnet of RE-TM. The temperature dependence of the critical field is well described in the framework of the theory of the molecular field. The value of the effective constant of the intersublattice exchange interaction turned out to be comparable to  $\lambda$  for a single crystal of DyCo<sub>5.3</sub> and several times higher than the same value for Dy-Co films obtained by thermal sputtering.

### Funding

The results were obtained as part of implementation of the state assignment of the Ministry of Education and Science of Russia FEUZ-2023-0020.

### **Conflict of interest**

The authors declare that they have no conflict of interest.

### References

- K. Vahaplar, A.M. Kalashnikova, A.V. Kimel, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing. Phys. Rev. Lett. 103, 117201 (2009).
- [2] Z. Hu, J. Besbas, R. Smith, N. Teichert, G. Atcheson, K. Rode, P. Stamenov, J.M.D. Coey. Appl. Phys. Lett. 120, 112401 (2022).
- [3] J. Becker, A. Tsukamoto, A. Kirilyuk, J.C. Maan, Th. Rasing, P.C.M. Christianen, A.V. Kimel. Phys. Rev. Lett. 118, 117203 (2017).
- [4] V.O. Vaskovsky, E.V. Kudyukov, E.A. Stepanova, E.A. Kravtsov, O.A. Adanakova, A.S. Rusalina, K.G. Balymov, A.V. Svalov. FMM 122, 5, 513 (2021). (in Russian).
- [5] B.P. Khrustalev, V.G. Pozdnyakov, G.I. Frolov, V.Yu. Yakovchuk. FTT **31**, *3*, 112 (1989). (in Russian).
- [6] R.S. Srivastava. J. Appl. Phys. 48, 1355 (1977).
- [7] V.O. Vaskovsky, K.G. Balymov, A.V. Svalov, N.A. Kulesh, E.A. Stepanova, A.N. Sorokin. FTT 53, 11, 2161 (2011). (in Russian).

- [8] A.G. Berezin, R.Z. Levitin, Yu.F. Popov. ZhETF **79**, 268 (1980). (in Russian).
  - [9] R. Hasegawa. J. Appl. Phys. 46, 5263 (1975).
  - [10] A. Gangulee, R.J. Kobliska. J. Appl. Phys. 49, 4896 (1978).
  - [11] P. Hansen, C. Clausen, G. Much, M. Rosenkranz, K. Witter. J. Appl. Phys. 66, 756 (1989).
- [12] A.V. Svalov, O.A. Adanakova, V.O. Vas'kovskiy, K.G. Balymov, A. Larrañaga, G.V. Kurlyandskaya, R. Domingues Della Pace, C.C. Plá Cid. J.M. Magn. Mater. 459, 57 (2018).
- [13] T.-h. Wu, H. Fu, R.A. Hajjar, T. Suzuki, M. Mansuripur. J. Appl. Phys. 73, 1368 (1993).
- [14] G.I. Frolov, V.S. Zhigalov. Fizicheskie svojstva i primenenie magnitoplenochnyh nanokompozitov. Izd-vo SO RAN, Novosibirsk (2006). 187 s. (in Russian).
- [15] A.S. Rusalina, V.N. Lepalovsky, E.A. Stepanova, V.O. Vaskovsky, G.V. Kurlandskaya, A.V. Svalov. FTT 65, 6, 883 (2023).

Translated by A.Akhtyamov