### 01,05,13

# Anomalous hysteresis loops of ferrimagnetic Gd-Co films of different thickness near the magnetic compensation temperature

© A.S. Rusalina<sup>1</sup>, V.N. Lepalovskij<sup>1</sup>, E.A. Stepanova<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 17, 2023 Revised April 17, 2023 Accepted May 11, 2023

Triple hysteresis loops were observed for amorphous ferrimagnetic Gd-Co films near the magnetic compensation temperature, which can be a consequence of both the spin-flop transition and the chemical composition gradient. In the work, an assessment of the possible chemical inhomogeneity of the films based on magnetic measurements was carried out and its relationship with the thickness of the samples was observed.

Keywords: amorphous magnetic films, perpendicular magnetic anisotropy, ferrimagnetism, magnetic domain structure, magnetic hysteresis.

DOI: 10.21883/PSS.2023.06.56089.08H

## 1. Introduction

Near the compensation temperature  $T_{\rm comp}$  in a ferrimagnetic, a spin-flop transition can be realized under the influence of an external magnetic field disrupting the antiparallel ordering of the sublattice magnetizations and results in the so-called noncollinear magnetic state [1]. For rare-earth ---transition metal (RE-TM) alloy films, this opens up the additional possibility of using them as material for noncollinear spintronics [2]. Generally, the presence of a spin-flop transition is captured through the appearance of specific features in the area of large fields on magnetometric, magneto-optical and magnetoresistive hysteresis loops [3–7]. At the same time, as shown earlier, the unusual triple hysteresis loops of ferrimagnetic RE-TM films may be a consequence of chemical heterogeneity of samples [8-10]. A model has been proposed to describe such films remagnetization by splitting an inhomogeneous sample into two exchange-bonded layers of different averaged chemical composition, separated by a compensating surface, near which the film magnetization at a fixed temperature tends to zero [8,11]. This paper presents the results of a study of the magnetic properties of amorphous ferrimagnetic films Gd-Co over a wide temperature range, including the compensation temperature, while varying the thickness films.

## 2. Technique of studies

 $Gd_{21.6}Co_{78.4}$  films were deposited on glass substrates by magnetron sputtering of Gd and Co targets in an argon atmosphere. The film thickness ranged from 10 to 100 nm. All samples were protected against oxidation by buffer and covering Ta layers 5 nm thick. The magnetic properties were studied with a MPMS-7XL measuring complex in the temperature range of 5 to 300 K and an Evico magneto-optical Kerr microscope at room temperature.

# 3. Produced findings and discussion

The shape of the magnetometric hysteresis loops measured at both field orientations along and perpendicular to the sample plane indicates that the 10, 20 and 40 nm thick films exhibited perpendicular magnetic anisotropy throughout the temperature range studied. The compensation temperature of the films was determined as the value corresponding to a minimum on the temperature dependence of the magnetization M(T) (Fig. 1).

For films 20 and 40 nm thick,  $T_{\rm comp}$  was approximately 220 K. For a 10 nm thick sample,  $T_{\rm comp}$  was much lower. This is most likely due to the size factor as well as the possible influence on the electronic structure of the cobalt atoms of the conduction electrons of the Ta [12,13] protective layers, which ultimately results in the "magnetic active" composition of the Gd-Co(10 nm) layer, determining the value $T_{\rm comp}$ , being different from its chemical composition.

The magnetometric hysteresis loops measured on 20 and 40 nm thick films exhibited breaks characteristic of the spinflop transition at temperatures in the vicinity of  $T_{\text{comp}}$ . As an example, Fig. 2, *b* shows the loops for a sample of thickness 20 nm, with arrows indicating the characteristic breaks. For



**Figure 1.** Temperature dependences of magnetization of films Gd-Co of different thicknesses measured in a 10 kOe field oriented perpendicular to the sample plane.

100 nm thick films, the critical field of additional hysteresis loops  $H_{cr}$  is markedly smaller than for 20 and 40 nm thick films, and the shape of the loops is more characteristic of an exchange-coupled bilayer system (Fig. 2, c). The analogous hysteresis loops for the thinnest film do not show any characteristic breaks (Fig. 2, a), possibly due to the small signal from the sample.

Note that for films of all thicknesses, the features were found not only in the hysteresis loops but also in the temperature dependences of magnetization measured at field values greater than 10 kOe. As an example, Fig. 3 shows a set of such M(T) dependences for a 40 thick sample. It can be seen that the dependences M(T)measured at H equal to 30, 50 and 70 kOe show a deviation from the linear path around  $T_{\rm comp}$ , with the width of this temperature interval  $\Delta T$  increasing with the growth of *H*. Similar features were recorded in M(T) dependencies even for 10 nm thick film. Like the characteristic breaks in hysteresis loops, deviations from the linear course of the dependencies can be caused by spin-flop transitions or by heterogeneities in the chemical composition of the sample. Assuming the presence of chemical heterogeneity in the sample, then the increase  $\Delta T$ with increasing H can be explained by assuming, by analogy with models [9,11], that a compensating surface exists in the studied film, which divides the sample into two layers. The total magnetic moment of one  $M_1$  layer is determined by the predominant moment of the rareearth sublattice, the moment of the other  $M_2$  layer is dominated by the transition metal sublattice. These layers are exchange-coupled, and the exchange energy is minimal when the magnetic moments of the layers are antiparallel. As the temperature changes, the compensation surface moves across the sample volume along the composition gradient. A relatively weak magnetic field cannot disturb the antiparallel arrangement of the magnetic moments of the layers, so that the sample behaves like a homogeneous film with a magnetic moment  $M = M_1 - M_2$  as the temperature changes. In a field larger than  $H_{cr}$ , at a certain temperature

above the average compensation temperature of one of the layers, an increase in the Zeeman energy of this layer causes its magnetic moment to reorient, thus increasing



**Figure 2.** Magnetic hysteresis loops measured at temperatures near  $T_{\text{comp}}$  for samples of thicknesses 10 nm(a), 20 nm(b) and 100 nm(c).



**Figure 3.** Temperature dependences of the magnetization of a 40 nm thick Gd-Co film measured in a field of different strengths oriented perpendicular to the sample plane. The value of the temperature interval  $\Delta T$ , in which the deviation from the linear course of the M(T) dependence is observed, is shown for H = 70 kOe.



**Figure 4.** Dependence of the temperature interval  $\Delta T$  on the measuring field value *H* for an Gd-Co film 10 nm thick.

the total magnetic moment of the sample. In this case, a magnetic inhomogeneity like a domain boundary occurs in the vicinity of the compensation surface. As the temperature increases, the volume of the second layer and its energy in the external field decrease. Therefore, at a certain temperature, slightly lower than the average compensation temperature of the second layer, the opposite field orientation of the magnetic moment of this layer becomes advantageous due to a gain in the exchange energy of the sample, i.e. the magnetic moment of the second layer is reoriented. This is accompanied by the disappearance of magnetic inhomogeneity at the layer boundaries. It is clear, that increasing the measurement

field causes a remagnetization of layers with a smaller volume and lower average magnetization, resulting in a higher  $\Delta T$  interval and allowing less heterogeneity in the chemical composition of the film to be sensed. In order to make a simple quantification of the possible heterogeneity of the film Gd-Co, we will assume that the sample has a constant thickness gradient of chemical composition, and the minimum field value, at which one of the layers is reoriented, is  $H_{\rm cr}$ . At moment of remagnetization, the Zeeman energy of the given layer is equal to the energy of the resulting magnetic inhomogeneity of the domain boundary type, where  $\gamma = M_1 H l$ , where  $\gamma$  — boundary energy density,  $M_1$  — average magnetization of the layer, H external field, l — layer thickness. The boundary energy density can also be written as  $\gamma = 4\sqrt{AK}$ , where A the exchange interaction parameter, K — the anisotropy constant. In amorphous Gd-Co films, these constants have the following orders of magnitude:  $A = 1 \cdot 10^{-6} \text{ erg/cm}$ ,  $K = 1 \cdot 10^5 \text{ erg/cm}^3$ , so,  $\gamma \approx 1.3 \text{ erg/cm}^2$ . The value of  $H_{\rm cr}$ is determined by linear extrapolation of the dependence  $\Delta T(H)$  obtained from the set of M(T, H) dependences as shown in Fig. 4 for a sample thickness of 10 nm. Values  $H_{cr}$  for the 10, 20 and 40 nm samples were 15, 15 and 10 kOe, respectively. If we take half of the film thickness, i.e. 5 nm for a 10 nm thick sample, and substitute the corresponding values in the formula  $\gamma = M_1 H l$ , we obtain  $M_1 \approx 170$  Gs. Assuming a constant composition gradient across the sample thickness, we obtain that the change in magnetization in the layer  $\Delta M_1 = 2M_1$ , and in the whole sample  $\Delta M = 2\Delta M_1 \approx 700 \,\text{Gs.}$  Using the ratio  $\Delta x = 1.25 \cdot 10^{-4} \Delta M$  found in [11], we obtain that the change in chemical composition across the sample thickness  $\Delta x$  is 8 at.%. Similar procedures for samples 20 nm and 40 nm thick give a value  $\Delta x$  of 4 at.%. and 3 at.%, respectively. The unreasonably high values  $\Delta x$ , especially compared to  $\Delta x = 0.5$  at.% obtained for Gd-Co film of  $1 \mu m$  thickness deposited under similar conditions [14]. Furthermore, it seems illogical that  $\Delta x$  decreases with increasing film thickness. Thus, our assumption that the anomalies in the temperature dependences M(T)measured in large fields are due to heterogeneity in the chemical composition of these samples is not conclusively confirmed.

The observed  $H_{cr}$  for our samples are in agreement with the values of spin-flop transition fields available in the literature for amorphous RE-TM [4–7] films, and are several tens kOe, which is an order of magnitude smaller than the  $H_{cr}$  obtained for single-crystal GdCo<sub>5</sub>(460 kOe) [3].

Value  $\Delta x$  based on the above methodology for a 100 nm thick film could not be estimated, since it was impossible to determine  $H_{cr}$  from the set of dependences M(T), as these dependences exhibit a non-linear character over a wide temperature range even in relatively weak fields (Fig. 1). The magneto-optical hysteresis loops measured on this sample on the film side and the glass substrate side differ strongly from each other (Fig. 5), indicating heterogeneous chemical composition across thickness. Thus,



Figure 5. Magneto-optical hysteresis loops for a 100 nm thick sample, measured on the film side (a) and on the substrate side (b).

the triple hysteresis loops for this sample (Fig. 2, c) are due to its chemical heterogeneity.

# 4. Conclusion

The anomalous triple hysteresis loops observed in the studied 100 nm thick films  $Gd_{21.6}Co_{78.4}$  are due to the presence of a chemical composition gradient across the sample thickness. The remagnetization of such a film can be described within the previously proposed model [9,11], assuming the existence of a compensating surface in the film that dividing the sample into two exchange-coupled layers. In smaller films, the inhomogeneity of the chemical composition is not sufficient for a two-layer model, and the anomalous hysteresis loops are most likely due to spin-flop transitions in strong fields. However, more complex chemical fluctuations observed in amorphous RE-TM [15,16] films cannot be excluded as a possible cause of the anomalous loops.

#### 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

[1] A.E. Clark, E. Callen. J. Appl. Phys. 39, 5972 (1968).

- [2] M.B. Jungfleisch, W. Zhang, A. Hoffmann. Phys. Lett. A 382, 865 (2018).
- [3] Yu. Skourski, M.D. Kuz'min, K.P. Skokov, M. Richter, D. Eckert, I.S. Tereshina, K.-H. Müller. J. Magn. Magn. Mater. 290–291, 435 (2005).
- [4] Becker, A. Tsukamoto, A. Kirilyuk, J.C. Maan, Th. Rasing, P.C.M. Christianen, A.V. Kimel. Phys. Rev. Lett. 118, 117203 (2017).
- [5] M.D. Davydova, K.A. Zvezdin, J. Becker, A.V. Kimel, A.K. Zvezdin. Phys. Rev. B 100, 064409 (2019).
- [6] T. Fu, S. Li, X. Feng, Y. Cui, J. Yao, B. Wang, J. Cao, Z. Shi, D. Xue, X. Fan. Phys. Rev. B 103, 064432 (2021).
- [7] D. Chen, Y. Xu, S. Tong, W. Zheng, Y. Sun, J. Lu, N. Lei, D. Wei, J. Zhao. Phys. Rev. Mater. 6, 014402 (2022).
- [8] M. Amatsu, S. Honda, T. Kusuda. IEEE Trans. Magn. 13, 1612 (1977).
- [9] R.I. Tagirov, A.A. Glaser. FMM 46, 1, 75 (1978). (in Russian).
- [10] T. Kobayashi, H. Tsuji, S. Tsunashima, S. Uchiyama. Jpn. J. Appl. Phys. 20, 11, 2089 (1981).
- [11] V.O. Vaskovsky, V.V. Lesnykh, G.S. Kandaurova, T.H. Agamalian. FMM 59, 3, 470 (1985). (in Russian).
- [12] A.V. Svalov, V.O. Vas'kovskiy, G.V. Kurlyandskaya. Phys. Met. Metallogr. 118, 13, 1263 (2017).
- [13] 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. Magn. Magn. Mater. 459, 57 (2018).
- [14] A.N. Sorokin, A.V. Svalov. Vacuum 46, 2, 1131995 (1995).
- [15] K. Chen, D. Lott, F. Radu, F. Choueikani, E. Otero, Ph. Ohresser. Sci. Rep. 5, 18377 (2016).
- [16] J.-L. Bello, D. Lacour, S. Migot, J. Ghanbaja, S. Mangin, M. Hehn. Appl. Phys. Lett. **121**, 212402 (2022).

Translated by Ego Translating