Наносистеми, наноматеріали, нанотехнології
Nanosistemi, Nanomateriali, Nanotehnologii
2024, т. 22, № 4, сс. 785–791
https://doi.org/10.15407/nnn.22.04.785

PACS numbers: 68.37.Hk, 73.40.Gk, 73.43.Qt, 75.47.-m, 75.50.Tt, 75.70.-i, 81.40.Rs

The Estimating Method for the Enhancement of the Magnetization in the Fe/Gd_2O_3 Nanostructure Resulting from the *f*–*d*-Exchange Interaction

A. M. Kasumov, A. I. Dmitriev, K. A. Korotkov, V. M. Karavaieva, and A. I. Ievtushenko

I. M. Frantsevich Institute for Problems in Materials Sciences, N.A.S. of Ukraine,
3, Omeljan Pritsak Str.,
03142 Kyiv, Ukraine

The tunnelling magnetoresistance (*TMR*) effect in Fe/Gd_2O_3 nanostructure consisting of Fe island film deposited on Gd_2O_3 layer is investigated. In Fe, Co, Ni/rare earth metals (REM) oxide nanostructures, an exchange *f*–*d* interaction occurs in the region of their interface due to wave-functions' hybridization of *f*- and *d*-electrons with unfilled shells. This leads to an additional magnetization M_{f-d} and effective magnetic field H_{f-d} corresponding to it. The practical value of this enhancement of magnetic properties is that it is achieved in a nanoscale volume without the use of energy or amplification devices. For nanotechnology applications, this fact is important. A method is proposed to determine H_{f-d} and M_{f-d} based on the study of *TMR*(**H**) dependence, information about which is lacking. The hybridization propagation region in the Fe/Gd_2O_3 structure is shown to extend at least 70 nm deep in the REM oxide layer.

Досліджено ефект тунельного магнетоопору (*TMR*) в наноструктурі Fe/Gd_2O_3, що складається з острівцевої плівки Fe, нанесеної на шар Gd_2O_3. В наноструктурах оксидів Fe, Co, Ni/рідкісноземельних металів (РЗМ) в області їхньої межі поділу виникає обмінна *f*–*d*-взаємодія, зумовлена гібридизацією хвильових функцій *f*- і *d*-електронів з незаповненими оболонками. Це приводить до додаткової намагнетованости M_{f-d} і відповідного їй ефективного магнетного поля H_{f-d}. Практична цінність такого посилення магнетних властивостей полягає в тому, що воно досягається в нанорозмірному об'ємі без використання енергії або підсилювальних пристроїв. Для нанотехнологічних застосувань цей факт є важливим. Запропоновано метод визначення H_{f-d} і M_{f-d} на основі дослідження залежности *TMR*(**H**), інформація про яку відсутня. Показано, що область поширення гібридизації в структурі Fe/Gd_2O_3 простягається щонайменше на 70 нм углиб оксидного шару РЗМ.

Key words: tunnelling magnetoresistance, Fe, rare earth metals' oxide, exchange f–d interaction.

Ключові слова: тунельний магнетоопір, Fe, оксид рідкісноземельних металів, обмінна f–d взаємодія.

(Received 8 August, 2023)

1. INTRODUCTION

In nanostructures consisting of films of the iron group metals Fe, Co, Ni and rare earth metal (REM) oxides, f–d-exchange interaction occurs between atoms with unfilled f- and d-electron shells contained in the film in the region of their interface [1].

It has been shown by the anomalous Hall effect method [2] that such exchange f–d interaction leads to an increase of their magnetization by the value of M_{f-d}. In this case, many properties of the nanostructure related to magnetization are enhanced: magnetoresistance [3], Faraday effect [4], electron paramagnetic resonance [5], conductivity of MDM structures [6], and tunnelling magnetoresistance. The practical value of this property enhancement is that it is achieved at the nanoscale without energy consumption and without the use of amplifying devices. This fact is important for the application of nanotechnology.

The aim of the present work is the estimation of the magnitude of the effective magnetic field H_{f-d} and the magnetization M_{f-d} in Fe, Co, Ni/REM oxide nanostructures, for which no information is available.

In order to achieve this goal, a Fe/Gd_2O_3 nanostructure consisting of a Fe island film deposited on a Gd_2O_3 layer has been selected as the object of study. These materials were chosen because both Fe in the series of the iron group and Gd in the series of the lanthanides possess one of the largest effective magnetic moments $\mu_{Fe} = 7.13\mu_B$ and $\mu_{Gd} = 7.95\mu_B$ [7]. There is a high energy of exchange f–d interaction arising between islands of Fe film and the layer of Gd_2O_3, on which they are located, according to the theory [8].

The conductivity in island films is due to tunnelling transitions of electrons through small gaps of a few angstroms that separate the islands from each other [9]. A tunnelling magnetoresistance (*TMR*) effect realized by the simultaneous influence of the exchange f–d interaction between Fe and Gd_2O_3 occurs when an external magnetic field **H** is applied to the island film. The peculiarities of the *TMR*(**H**) field dependence under these conditions allow estimating H_{f-d} and M_{f-d}, as well as their dependence on structural layer thickness.

2. EXPERIMENTAL ENVIRONMENTS

The Fe and Gd_2O_3 films were deposited on a glass substrate by electron beam evaporation of targets with similar composition from chemically pure reactants. The deposition conditions were: Fe—vacuum $p = 3 \cdot 10^{-3}$ Pa, film growth rate $\upsilon = (5/10)$ nm/min, substrate temperature $t_{Sub} = (30/40)$°C; Gd_2O_3—O_2 partial pressure $p_{O_2} = 2 \cdot 10^{-2}$ Pa, film growth rate $\upsilon = (3/10)$ nm/min, substrate temperature $t_{Sub} = (30/50)$°C. The Fe/Gd_2O_3 film structures were prepared by sequentially depositing Gd_2O_3 and then Fe layer on the substrate. The Fe island layer thickness was of 30 nm for all samples, while Gd_2O_3 thickness varied between 45 nm and 70 nm. For comparison, a similarly thick Fe film was deposited on a glass substrate without Gd_2O_3. The ends of the Fe films were coated with Cu electrodes by thermal evaporation for soldering conductors.

To stabilize the resistivity of the Fe island films, which increases due to oxidation in air, the samples were kept in the atmosphere at room temperature for 3 days. The Fe islands were coated with a thin layer of oxide preventing further oxidation and resistance change, which allowed *TMR* measurement. The surface oxidation of Fe islands did not pose a threat to the achievement of the goal, since we have previously shown in Ref. [1] that the exchange f–d interaction occurs not only at the interface of oxides with Fe, but also with Fe_3O_4. Magnetite Fe_3O_4 is a semi-metal due to its conductivity. It does not prevent the tunnelling of electrons between islands.

3. RESULTS AND DISCUSSION

3.1. Morphology of the Fe/Gd_2O_3 Nanostructure

Figure 1 shows an electron-microscope image of the Fe film (30 nm) on a layer of Gd_2O_3 (45 nm). It also shows an electronogram of this nanostructure. The image was acquired on a Tescan Mira SLMU scanning electron microscope.

The morphology of the Fe film corresponds to that of the island film. The islands have an asymmetric shape, apparently, due to oxidation in air. The average size of the islets ranges from 20–100 nm. The surface of the islands is inhomogeneous with cluster nanostructure.

Electronogram analysis shows that Fe film is polycrystalline, and the Gd_2O_3 film is amorphous. Electron phase analysis shows that 3 strong Fe lines each $d_1 = 2.0268$ Å, $d_2 = 1.4221$ Å, $d_3 = 1.1702$ Å [10] and Fe_3O_4 $d_1 = 2.967$ Å, $d_2 = 2.532$ Å, $d_3 = 1.616$ Å are present in the diagram [11]. Therefore, we can assume that the most likely model

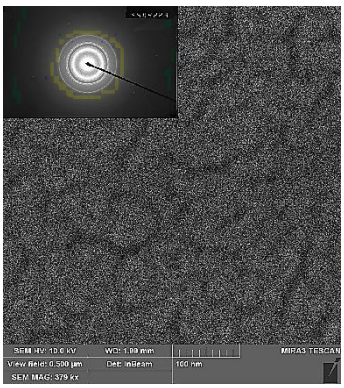

Fig. 1. Electron-microscopy image of a Fe film (30 nm) on a layer of Gd_2O_3 (45 nm) and an electronogram of this nanostructure.

for air-exposed iron islands is the core–shell configuration, where the core is Fe and the shell is Fe_3O_4 iron oxide. The average size of the coherent scattering regions of the Fe film determined from the electronogram by the Selyakov–Scherrer method is of 9.4 nm.

3.2. Estimated Value of the H_{f-d} and M_{f-d}

The *TMR* of Fe films deposited without and with Gd_2O_3 sublayer was calculated using the formula [9]

$$TMR = \frac{R(\mathbf{H}) - R(\mathbf{0})}{R(\mathbf{0})}, \qquad (1)$$

where $R(\mathbf{0})$ and $R(\mathbf{H})$ are resistances of the film without and under the influence of the external magnetic field **H**. The vector **H** was chosen perpendicular to both the Fe films surface and the vector of the current flowing through them.

Figure 2 shows the $TMR(H)$ dependence of 30 nm thick Fe island films deposited on a substrate without (curve *1*) and with (curve *2*) a Gd_2O_3 sublayer.

The $TMR(\mathbf{H})$ dependence for the two samples has some common features. First, *TMR* is a negative value, which is due to the influence of the magnetic field on the orientation of the electron spins in the islands: as *H* increases, more and more spins are oriented along the field direction, which facilitates the tunnelling of electrons between the islands without expending energy for spin rotation. In this case, the charge-transfer resistance decreases, and the *TMR* becomes negative.

The second feature is the appearance of a saturation region at

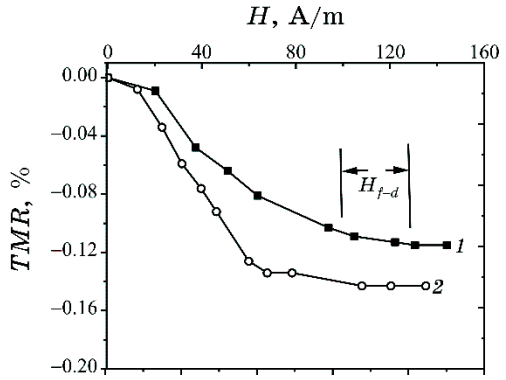

Fig. 2. *TMR* dependence on strength *H* for Fe (30 nm) (curve *1*) and Fe (30 nm)/Gd_2O_3 (70 nm) (curve *2*).

high field strength, which is a result of the all electron-spins' orientation in the islands along the direction of the vector **H**. At this point, the *TMR* no longer depends on the field strength *H*. For the Fe film on glass without a Gd_2O_3 sublayer (Fig. 2, curve *1*), the application of a much higher field strength $H_{\mathrm{Fe}}^{Sat} = 130.62$ A/m than for the Fe film on Gd_2O_3 (Fig. 2, curve *2*) $H_{\mathrm{Fe/Gd_2O_3}}^{Sat} = 108.02$ A/m, where H_{Fe}^{Sat} and $H_{\mathrm{Gd_2O_3}}^{Sat}$ are the external magnetic-field strengths required to reach the saturation region of the *TMR*(*H*) curve for Fe films without and with Gd_2O_3 sublayer, respectively. The reason for this difference cannot be due to the shielding of the strong magnetic field by the Gd_2O_3 layer, since it is paramagnetic with a relatively low value of magnetic permeability. The most likely reason for the decrease in the *H* value required to reach the *TMR* saturation state in the Fe/Gd_2O_3 structure (Fig. 2, curve *2*) compared to Fe without Gd_2O_3 (Fig. 2, curve *1*) could be the influence of the exchange *f*–*d* interaction between Fe and Gd_2O_3. This interaction leading to the appearance of an additional magnetization M_{f-d} and its corresponding effective field H_{f-d} should facilitate the achievement of the saturation state *TMR*(*H*) in Fe films due to the joint influence of the external field **H** and the effective magnetic field H_{f-d} on the Fe/Gd_2O_3 structure. In this case, the value of H_{f-d} is the difference between the saturation field values obtained in Fe films without and with the Gd_2O_3 layer (as shown in Fig. 2), *i.e.*,

$$H_{f-d} = H_{\mathrm{Fe}}^{Sat} - H_{\mathrm{Fe/Gd_2O_3}}^{Sat}, \qquad (2)$$

The obtained expression (2) can be used to determine the value of H_{f-d} based on the experimental *TMR*(*H*) dependence. In our case, $H_{f-d} = 22.6$ A/m.

Figure 3 shows the results of H_{f-d} calculations for the Fe/Gd_2O_3

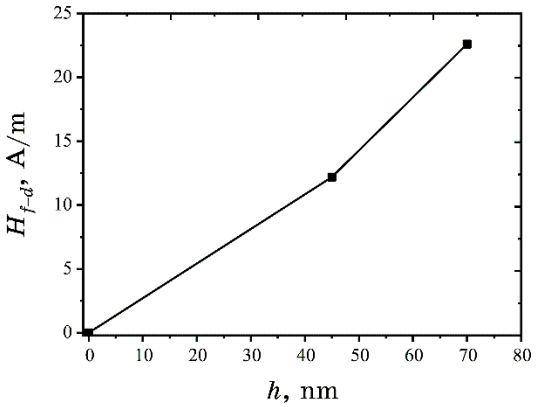

Fig. 3. Dependence of H_{f-d} on the thickness h of Gd_2O_3 film in Fe/Gd_2O_3 nanostructure.

structure as a function of Gd_2O_3 thickness.

As can be seen (Fig. 3), as the thickness of the Gd_2O_3 layer increases from 0 to 70 nm, the value of H_{f-d} reaches a value of 22.6 A/m, which is 20.9% of the saturation field value of 108 A/m in the Fe (30 nm)/Gd_2O_3 (70 nm) structure.

The effective magnetic field H_{f-d} is related to the magnetic flux M_{f-d} by the equation

$$H_{f-d} = \chi M_{f-d}, \tag{3}$$

where χ is the magnetic permeability as a function of the effective magnetic field strength. For the Fe (30 nm)/Gd_2O_3 (70 nm) structure, $M_{f-d} = 3.4 \cdot 10^5$ A/m. In estimating the value of M_{f-d}, it was assumed that the nature of the χ field dependence for the Fe (30 nm)/Gd_2O_3 (70 nm) structure is similar to the χ dependence of iron-ARMCO (technically pure iron) and insignificantly different in magnitude.

The influence of the exchange f–d interaction extends in Gd_2O_3 oxide to a significant distance ≥ 70 nm from the Fe interface. Large distances have been observed in other REM oxides too, while investigating other magnetic properties by nanostructures consisting of films of the iron-group metals (Fe, Co, Ni) and rare earth oxides [1].

4. CONCLUSIONS

A method is proposed to determine the effective magnetic-field strength H_{f-d} and the magnetization value M_{f-d} caused by the exchange f–d interaction. In Fe, Co, Ni/REM oxide nanostructures in

their interface region, an exchange f–d interaction occurs because of hybridization of wave functions of f- and d-electrons with unfilled shells. This results in an additional magnetization M_{f-d} and the corresponding effective field H_{f-d}. The method allows the determination of H_{f-d} and M_{f-d} and their dependence on the experimental conditions based on the TMR**(H)** study.

It is shown that the hybridization-propagation region in the Fe (30 nm)/Gd_2O_3 (70 nm) structure extends to a depth of at least 70 nm in the REM oxide layer. The maximum values of H_{f-d} and M_{f-d} obtained for Fe (30 nm)/Gd_2O_3 (70 nm) nanostructure are of 22.6 A/m and $3.4{\cdot}10^5$ A/m, respectively.

REFERENCES

1. A. M. Kasumov, A. I. Dmitriev, K. A. Korotkov, V. M Karavaeva, K. O. Vyshnevska, and A. I. Ievtushenko, *Chemistry, Physics and Technology of Surface*, **13**, No. 4: 434 (2022); https://doi.org/10.15407/hftp13.04.434
2. A. M. Kasumov, A. I. Dmitriev, M. V. Radchenko, A. E. Baybara, O. I. Bykov, K. A. Korotkov, V. M. Karavaeva, K. O. Vyshnevska, O. I. Olifan, and A. I. Ievtushenko, *Chemistry, Physics and Technology of Surface*, **13**, No. 1: 105 (2022); https://doi.org/10.15407/hftp13.01.105
3. A. M. Kasumov, V. M. Karavaeva, A. A. Mikitchenko, K. O. Shapoval, M. A. Perepelitsa, and G. V. Lashkarev, *Powder Metall Met. Ceram.*, **57**: 325 (2018); https://doi.org/10.1007/s11106-018-9985-x
4. A. M. Kasumov, V. M. Karavayeva, K. O. Shapoval, and G. V. Lashkarov, *Nanosistemi, Nanomateriali, Nanotehnologii*, **16**, Iss. 1: 181 (2018); https://doi.org/10.15407/nnn.16.01.181
5. A. M. Kasumov, A. I. Dmitriev, Yu. M. Bataiev, M. M. Bataiev, V. M. Karavaeva, and K. A. Korotkov, A. I. Ievtushenko, *Chemistry, Physics and Technology of Surface*, **12**, No. 2: 144 (2021); https://doi.org/10.15407/hftp12.02.144
6. A. M. Kasumov, A. I. Dmitriev, Yu. M. Bataiev, M. M. Bataiev, V. M. Karavaeva, K. A. Korotkov, and A. I. Ievtushenko, *Chemistry, Physics and Technology of Surface*, **12**, No. 2: 144 (2021); https://doi.org/10.15407/hftp12.02.144
7. G. V. Samsonov, *Handbook of the Physicochemical Properties of the Elements* (New York: Springer: 1968); https://doi.org/10.1007/978-1-4684-6066-7)
8. G. T. Trammell, *Phys. Rev.*, **131**: 932 (1963); https://doi.org/10.1103/PhysRev.131.932
9. J. S. Moodera and G. Mathon, *J. Magn. Magn. Mater.*, **200**, Nos. 1–3: 248 (1999); https://doi.org/10.1016/S0304-8853(99)00515-6
10. *Powder Diffraction Files* (PDF [06-0696]). Powder Diffraction File™ (PDF®) Search; https://www.icdd.com/pdfsearch/
11. *Powder Diffraction Files* (PDF [19-629]). Powder Diffraction File™ (PDF®) Search; https://www.icdd.com/pdfsearch/