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Interfacial ferromagnetism in sputtered Pt/Gd/Pt system at room temperature

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Abstract. The magnetic properties of ultrathin Pt/Gd/Pt films prepared by magnetron sputtering were investigated. The Gd layer thickness varied from 1 to 15 nm. At room temperature, the samples exhibited a weak saturation magnetization, predominantly of interfacial origin. The measured interfacial magnetic moment was 14.5 $\mu ergG^{-1}cm^{-2}$. The contribution of bulk Gd layers to the saturation magnetization was found to be negligible. The shape of the hysteresis loops did not depend on the Gd layer thickness. Crystallization into a face-centered cubic structure was observed only for the 15 nm Gd layer. The results of this study emphasize the importance of interfacial ferromagnetism in the Pt/Gd/Pt system.

Keywords: gadolinium, superparamagnetism, multilayer films, magnetization, magnetic susceptibility, magnetic ordering temperature, size-dependent magnetic properties

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Материалы конференции
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Межфазный ферромагнетизм в напыленной системе Pt/Gd/Pt при комнатной температуре

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Аннотация. Исследованы магнитные свойства ультратонких пленок Pt/Gd/Pt, полученных методом магнетронного распыления. Толщина слоя Gd варьировалась от 1 до 15 нм. При комнатной температуре в образцах зафиксирован слабый магнитный момент насыщения, имеющий преимущественно интерфейсную природу. Измеренный интерфейсный магнитный момент составил 14,5 $мкЭргГс^{-1}см^{-2}$. Вклад в магнитный момент насыщения от объемных слоев Gd оказался пренебрежимо мал. Форма петель гистерезиса образцов не зависела от толщины слоя Gd. Кристаллизация в

гранецентрированную кубическую структуру наблюдается лишь при толщине Gd, равной 15 нм. Результаты проведенного исследования подчеркивают важность интерфейсного ферромагнетизма в системе Pt/Gd/Pt.

Ключевые слова: гадолиний, суперпарамагнетизм, многослойные пленки, намагниченность, магнитная восприимчивость, температура магнитного упорядочения, размерно-зависимые магнитные свойства

Финансирование: Высокоэффективное переключение намагниченности через спин орбитальный вращательный момент в магнитных наногетероструктурах для бесполевой SOT-MRAM памяти и устройств типа «вычисление в памяти», № 25-42-00083; Работа выполнена при поддержке Министерства науки и высшего образования Российской Федерации (Государственное задание №FZNS-2023-0012).

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Introduction

Gadolinium is a ferromagnetic material with a Curie temperature of 293.4 K in the bulk state [1]. In bulk samples and thick Gd films, a hexagonal close-packed (hcp) structure is formed. However, in ultrathin films, a face-centered cubic (fcc) phase is observed [2]. In films with a thickness of about 10 nm and an fcc structure, a Curie temperature above 300 K was recorded, and the saturation magnetization at 50 K reached approximately 100 $\mathrm{ergG^{-1}cm^{-3}}$.

Due to its unique magnetic properties, Gd is widely used in the creation of ferrimagnetic systems. For example, $\mathrm{Co}_x\mathrm{Gd}_{1-x}$ alloys exhibit ferrimagnetism [3]. The magnetic parameters of ferrimagnetic alloys, including the compensation temperature and saturation magnetization, can be precisely tuned by varying the alloy composition. In Pt/Co/Gd heterostructures, it has been established that the magnetic moments of ultrathin Gd layers deposited on Co are antiparallel to those of Co [4]. However, some studies, based on density functional theory calculations, have shown that in Pt/Co/Gd structures, only the magnetization of the two interfacial monolayers of Gd is oriented antiparallel to the magnetization in the Co layers, whereas the magnetization in the bulk Gd layers is aligned parallel to that in the Co layers [5].

In multilayer Pt/Co/Gd systems, the breaking of inversion symmetry along the film normal leads to the emergence of perpendicular magnetic anisotropy, spin–orbit torque [6], and the Dzyaloshinskii–Moriya interaction [7]. The presence of these effects in magnetic media is important for the development of information storage devices based on such media, for example, skyrmion-based memory [8]. To stabilize labyrinth and skyrmion magnetic configurations, superlattices of the form $[\mathrm{Pt/Co/Gd}]_n$ are employed [9]. It is believed that at room temperature the spontaneous magnetization in the Gd layers is caused by antiferromagnetic coupling with the magnetization of the Co layer, occurring at the Co/Gd interface.

In this work, the possibility of ferromagnetism arising solely from the Pt/Gd and Gd/Pt interfaces is investigated in a symmetric trilayer Pt/Gd/Pt system fabricated by magnetron sputtering. The aim of the study is to determine the crystal structure and magnetic properties of the Pt/Gd/Pt system while varying the thickness of the Gd layer.

Materials and Methods

The samples were fabricated by magnetron sputtering onto naturally oxidized silicon substrates $\mathrm{Si/SiO_2}$. Deposition was carried out in an Omicron vacuum chamber equipped with a substrate holder rotating at 40 revolutions per minute and four magnetron targets, ensuring uniform deposition of different materials without breaking the vacuum environment.

The thickness of the deposited layers was monitored using a quartz crystal thickness monitor with an accuracy of 0.1 nm. The sample structure was Pt (3 nm)/Gd (1−15 nm)/Pt (3 nm). For brevity, a sample with a Gd layer thickness of X nm will hereafter be denoted as Gd(X).

Deposition was carried out in an argon atmosphere at a base pressure of $P_0 = 1\times10^{-8}$ Torr and a working pressure of 1×10^{-3} to 5×10^{-3} Torr at a temperature of 300 K.

Magnetic properties and anisotropy were investigated using vibrating sample magnetometry (VSM) at room and low temperatures. The work presents magnetic hysteresis loops obtained with a SQUID magnetometer. The sample structure was studied by X-ray diffraction (XRD).

Results and Discussion

Analysis of the XRD spectra shown in Figure 1 indicates that a polycrystalline fcc Gd structure with (222) texture forms only in the Gd(15) sample, whereas thinner Gd films remain amorphous.

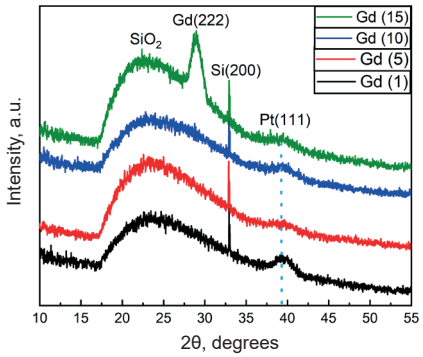

Fig. 1. XRD spectra of Pt/Gd/Pt samples with varying Gd layer thickness; formation of the fcc structure is observed only at a Gd thickness of 15 nm

The normalized magnetic hysteresis loops of the samples, measured at room temperature in magnetic fields oriented parallel (IP) and perpendicular (OOP) to the sample plane, are shown in Fig. 2, *a* and 2, *b*, respectively. The presented hysteresis loops were obtained after subtracting the linear signal from the nonmagnetic layers of the samples and the sample holder from the experimental loops.

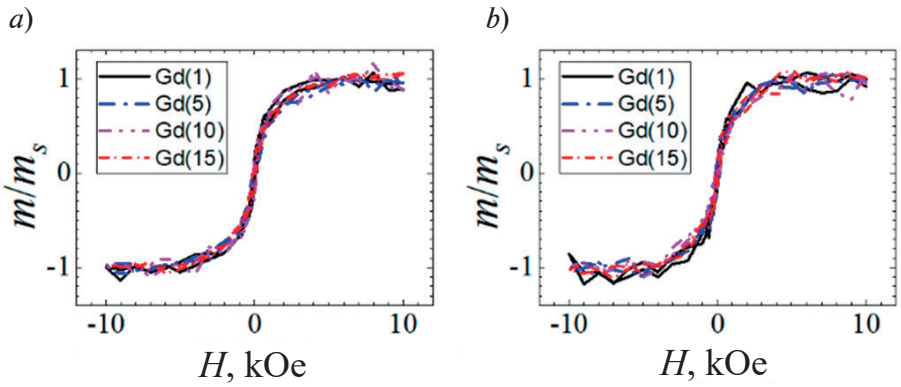

Fig. 2. Normalized magnetic hysteresis loops of the samples with the magnetic field oriented: in the film plane (IP) (*a*), perpendicular to the film plane (OOP) (*b*)

The hysteresis loops of all samples have a similar shape: they are isotropic and exhibit no hysteresis. Varying the Gd layer thickness from 1 to 15 nm does not affect the shape of the loops. Thus, it can be concluded that changes in the crystalline structure of Gd with increasing thickness do not significantly influence the magnetic properties of the system. The behavior of the samples resembles superparamagnetic systems composed of non-interacting magnetic nanoparticles or isolated magnetic crystalline grains with randomly oriented magnetic moments due to thermal fluctuations [10, 11, 12].

It is worth noting that hysteresis-free loops can be observed in Gd films at temperatures close to their Curie temperature [13]. The magnetic hysteresis loops of the samples measured by VSM were recorded at $T = 293$ K, which coincides with the Curie temperature of bulk Gd. However, in Gd films thinner than 5 nm, a reduction in the Curie temperature compared to bulk Gd is expected due to size effects [14, 15, 16]. In such a case, the hysteresis loop shape of the Gd(1) sample would significantly differ from those of samples with Gd thicknesses of 5 nm and above, which is not observed experimentally.

Fig. 3 shows the dependence of the saturation magnetic moment normalized to unit area on the Gd layer thickness. It is evident that the magnetic moment increases only slightly with increasing Gd thickness, indicating its interfacial origin. The dependence of the saturation magnetic moment, ms, on the thickness of the Gd magnetic layer, dGd, is described by the equation:

$$\frac{m_s}{S} = M_{s,\mathrm{V}} d_{\mathrm{Gd}} + \frac{m_{s,\mathrm{int.}}}{S}, \tag{1}$$

where $M_{s,\mathrm{V}}$ is the volumetric saturation magnetization of Gd, $m_{s,\mathrm{int.}}$ is the interfacial saturation magnetic moment of Gd, and S is the area of the magnetic film. Extrapolating the experimentally obtained graph to its intersection with the magnetic moment axis according to equation (1) yields the value of the interfacial magnetic moment normalized to unit area, $m_{s,\mathrm{int}}/S = 14.5$ μergG^{-1}cm^{-2}. Since approximately the same interfacial magnetic moment is observed in the Gd(1) sample with the smallest Gd thickness of 1 nm, it can be assumed that in all samples the interfacial saturation magnetic moment originates from Gd layers with a total thickness of about 1 nm. Then, the volumetric magnetization of these interfacial layers can be estimated as: $M_{s,\mathrm{int}} = m_{s,\mathrm{int}}/S/(1\ \mathrm{nm}) = 145$ ergG^{-1}cm^{-3}. The volumetric magnetization of the inner Gd layers in samples with Gd thickness greater than 1 nm, calculated according to equation (1) from the slope of the straight line shown in Fig. 3, is $M_{s,\mathrm{V}} = 3$ ergG^{-1}cm^{-3}.

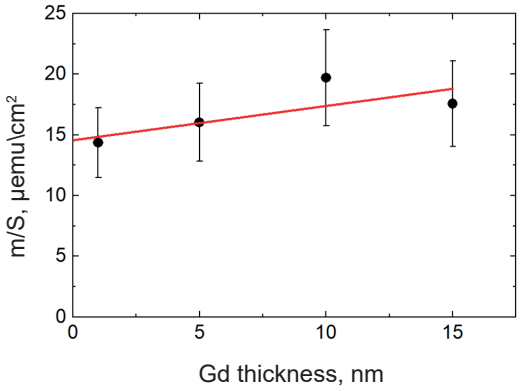

Fig. 3. Dependence of the saturation magnetic moment normalized to unit area on the thickness of the Gd layer; the solid line represents the linear approximation of the experimental data according to equation (1)

Temperature-dependent measurements of the saturation magnetic moment were performed for the series of samples (Fig. 4). The results show that the magnetic moment increases by only 10–20% compared to the value measured at room temperature when cooled down to 110 K. These findings support the conclusion that the absence of hysteresis and isotropy of the loops are not caused by the samples being close to their Curie temperature. Cooling the samples by 200 K below the Curie temperature would be expected to result in a significant increase in the saturation magnetic moment.

Interfacial superparamagnetism may be observed in Pt/Gd/Pt samples. Deposition by magnetron sputtering can sometimes cause intermixing of materials at the layer interfaces [17]. Further detailed investigation of the interface structure in this system is necessary. Nevertheless, assuming that the nonlinear magnetic signal arises from Gd magnetic clusters embedded in the Pt matrix, blocking of these nanoclusters can be expected at sufficiently low temperatures [10].

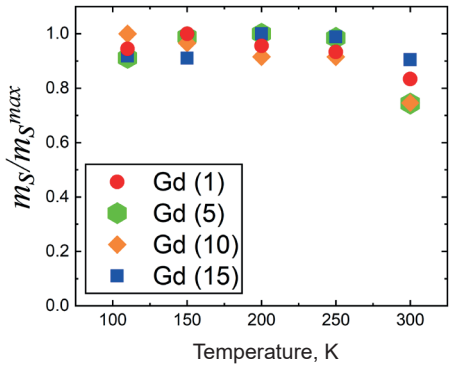

Fig. 4. Temperature dependence of the saturation magnetic moment (normalized to the maximum value in the temperature range) for samples with Gd layer thicknesses from 1 to 15 nm

Fig. 5 shows the magnetic hysteresis loops of the Gd(10) sample measured by SQUID magnetometry over the temperature range from 4 to 300 K. It can be seen that at 4 K the magnetization reversal occurs by a jump, while at other temperatures the reversal is gradual. Thus, the blocking temperature of this sample lies between 4 and 50 K. A more precise determination of the blocking temperature can be achieved by analyzing magnetization curves measured at different temperatures under different cooling conditions – with and without an applied magnetic field (FC – ZFC) [18].

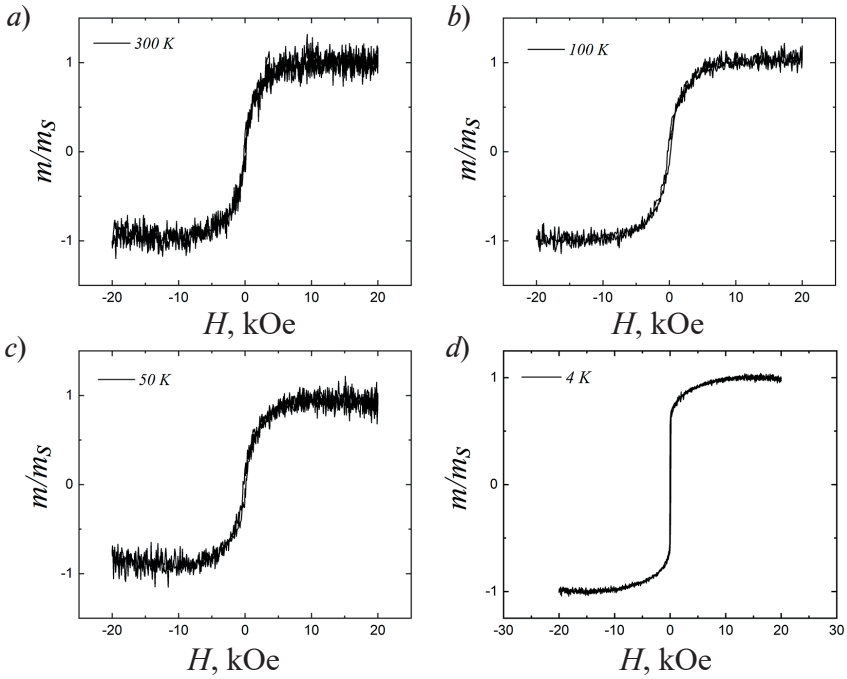

Fig. 5. Normalized magnetic hysteresis loops of the Gd(10) sample measured with the magnetic field oriented in the film plane at various temperatures: 300 K (*a*), 100 K (*b*), 50 K (*c*), 4 K (*d*)

In addition to the idea of interfacial superparamagnetism of Gd nanoclusters, the formation of a Pt-Gd alloy at the interfaces, exhibiting ferromagnetism at room temperature, can be considered to explain the observed results. However, no studies confirming this hypothesis have been found in the scientific literature. It is possible that the induced magnetization of Pt atoms due to the magnetic proximity effect plays some role in the observed ferromagnetism of the samples [19]. Notably, the magnetic proximity effect in Pt layers has been experimentally observed in systems where Pt layers form interfaces with alloys of transition and rare-earth metals. Moreover, the

direction of the induced magnetization in the Pt layers coincides with the magnetization direction in the transition metal layers, regardless of whether the magnetization of the sublattices of the transition or rare-earth metals predominates in the alloy [20]. Thus, the nature of the observed interfacial ferromagnetism in the Pt/Gd/Pt system remains unclear, and further studies are required to clarify it.

Conclusion

Isotropic superparamagnetic behavior independent of Gd layer thickness is observed in the symmetric trilayer Pt(3 nm)/Gd(1–15 nm)/Pt(3 nm) system. The saturation magnetization of the samples is predominantly due to interfacial effects, while the contribution from the bulk Gd layers is negligible. Films with Gd thicknesses below 10 nm remain amorphous, whereas a polycrystalline fcc phase forms at a Gd thickness of 15 nm. The interfacial magnetic moment value in the samples is 14.5 $\mu ergG^{-1}cm^{-2}$ at room temperature. The results demonstrate the possibility of realizing weak interfacial ferromagnetism of Gd adjacent to heavy metal Pt layers at room temperature.

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