Thin Solid Films 632 (2017) 79–87

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Microstructure of periodic metallic magnetic multilayer systems

Yu.M. Chesnokov [a], A.L. Vasiliev [a,b,*], G.V. Prutskov [a,c], E.M. Pashaev [a], I.A. Subbotin [a], E.A. Kravtsov [d,e], V.V. Ustinov [d]

[a] National Research Center "Kurchatov Institute", Ak. Kurchatov sq. 1, 123182 Moscow, Russia
[b] Insitute of Crystallography and Photonics RAS, Leninsky pr. 59, 119333 Moscow, Russia
[c] National Research University "Higher School of Economics", Myasnitskaya st. 20, 101000 Moscow, Russia
[d] Institute of Metal Physics, Ural Branch of the Russian Academy of Sciences, Sofia Kovalevskaya st. 18, 620990 Ekaterinburg, Russia
[e] Ural Federal University, Mira st. 2, 620002 Ekaterinburg, Russia

ARTICLE INFO

Article history:
Received 2 November 2016
Received in revised form 19 April 2017
Accepted 21 April 2017
Available online 26 April 2017

Keywords:
Metallic magnetic multilayers
Microstructure
Scanning transmission electron microscopy
Microanalysis
Resonant X-ray reflectivity

ABSTRACT

The results of a complex microstructural analysis of periodic metallic magnetic multilayer systems (MLS) by Scanning/Transmission Electron Microscopy, Energy Dispersive X-ray microanalysis and Resonant X-ray Reflectivity (RXRR) are presented. The crystal structure of MLS based on Cr-Gd-Cr-Fe layers with different Cr layer thicknesses was revealed. The use of RXRR at the Cr and Fe absorption edges significantly improves the optical contrast between correspondent layers. A significant Cr diffusion was detected in adjacent layers. After that diffusion, Fe and Gd layers transform to the solid solution of Cr in Fe and in Gd respectively, without changing their crystal structure. Such Cr behavior may affect both the exchange coupling between Fe and Gd magnetic moments and the structural parameters of Gd layers, thus influencing drastically the magnetic moment.

1. Introduction

Periodic magnetic metal multilayer systems (MLS), formed by alternating nanoscale layers of magnetic and non-magnetic metals are a relatively new class of artificial magnetic materials with unique physical properties that could be used for creating new devices for micro- and nanoelectronics [1]. So far, relatively well-studied magnetic metallic MLS are those based either on transition metals (TM) [2–3], or rare earth metals (REM) [4]. Recently, several research groups have started investigations of hybrid TM/REM systems, which are interesting as materials with a large magnetic moment [5,6]. Fe/Cr/Gd based periodic magnetic metal MLS is one of these systems in which originally antiferromagnetic interlayer exchange coupling of Fe—Gd can be modified by the introduction of an antiferromagnetic Cr spacer [5]. It was demonstrated that in a Fe/Cr/Gd three-layer system with 5 monolayers thickness of Cr layer, the ferromagnetic ordering of Fe and Gd magnetic moments occurs [5]. Moreover, the interlayer exchange interaction in Fe/Gd MLS oscillates depending on Cr layer thickness with a period of ~1.8 nm [7]. However, the total magnetic moment of the Fe/Cr/Gd MLS was found to be much less than expected for a multilayer with bulk-like magnetic moments in Gd and Fe layers [7–10]. The magnetic moment reduction in the multilayers is most likely due to their microstructure [6]. The imperfection of interfaces may affect both the exchange coupling between Fe and Gd magnetic moments mediated by a Cr layer, and structural parameters of Gd layers, influencing drastically the magnetic moment. It has been found recently that magnetic properties of metallic multilayers depend not only on layer thicknesses but also on interface roughness, defects, and crystallographic phases inside the layers [11]. In order to understand the physical mechanisms of these peculiarities in the magnetic properties, additional more precise experiments revealing the microstructural and chemical composition variation with atomic resolution of each layer and the interfaces between them are required.

Certainly, the usage of complementary methods in the analysis of MLS microstructure like the X-ray analysis and transmission electron microscopy (TEM), together with the analysis of the electrical parameters, provides more reliable information as has been shown in several studies, for example, in [12–17]. The scanning/transmission electron microscopy (S/TEM) as well as X-ray diffraction was used to analyze the interface structure and crystal lattice defects of $In_yAl_{1-y}As/In_xGa_{1-x}As$ heterostructures on InP substrates in [12]. The structural and magnetic properties of irradiated and non-irradiated MLS were studied in [13]. Combined electron microscopy and X-ray diagnostics data proved to be productive and useful in the study of multilayer heterostructures with layers of quantum dots [17] and Si-Ge heterostructures [18]. Such a combination of methods, which had not been explored in the investigations of these MLS before [7–9], was used in our study.

* Corresponding author.
E-mail address: AL_Vasilev@nrcki.ru (A.L. Vasiliev).

http://dx.doi.org/10.1016/j.tsf.2017.04.033
0040-6090/

2. Samples

The samples were a periodic system of alternating layers of [Fe/Cr/Gd/Cr] (repeated 12 times), grown with UHV magnetron sputtering onto the (001) and/or (111) Si substrate (Fig. 1). A preliminary study indicated that the orientation of the substrate does not affect MLS microstructure. Before growing, the substrate surfaces were cleaned chemically. The samples were grown at room temperature at the base pressure of 10^{-9} Torr. The growth started with a Cr buffer layer of about 5 nm in thickness, followed by a multilayered Fe/Cr/Gd structure. A protective Cr layer with the thickness of approximately 3 nm was deposited at the top of the MLS surface. All the samples differed in the thickness of Cr layers in the multilayer periodic systems. The scheme of MLS with an assumed thickness of the layers is presented in Fig. 1. A control sample was prepared without Cr layers, but only with alternating Fe and Gd layers.

3. Methods

3.1. Electron microscopy

The cross-section specimens for S/TEM were prepared using a focused ion beam (FIB) in a HeliosNanoLab ™ 600i scanning electron-ion microscope (FEI, USA) equipped with Pt and W gas injection systems (GIS) and with a Omniprobe 200 micromanipulator (Omniprobe, USA). To protect the sample surface during specimen preparation, a protective Pt layer with the thickness of 1.5 μm was deposited by an e^{-}- beam. A standard FIB procedure was used for S/TEM specimen preparation (see, for example [19]). The specimens were studied in a Titan 80-300 TEM/STEM (FEI, USA) with a spherical aberration (Cs probe) corrector at an accelerating voltage of 300 kV. The microscope was equipped with a field emission cathode (Schottky), a SuperTwin objective lens with the spherical aberration coefficient of 1.2 mm, an energy dispersive X-ray (EDX) spectrometer (EDAX, USA) and a high- angle annular dark-field (HAADF) electron detector (Fischione, USA). The EDX microanalysis including elemental mapping was additionally performed in a Tecnai Osiris TEM/STEM (FEI, USA) with an attached Super-X EDX system (Bruker, USA) at an accelerating voltage of 200 KeV. Digital Micrograph (Gatan, US) and TIA (FEI, US) software were used for image processing.

3.2. Resonant X-ray reflectivity (RXRR)

Several samples were studied by a high resolution RXRR method. The measurements were performed at the CuKα1 wavelength on a Rigaku SmartLab diffractometer (Rigaku, Japan) with a rotating Cu-anode. The experimental scheme included a 4xGe(220) Bartels monochromator, input and output collimation slits and a scintillation detector. The refractive index was $n = 1\text{-}\delta + i\beta$ (where δ and β were the refractive and absorption coefficients, respectively) for Gd for the CuKα1 wavelength differs substantially from Fe and Cr, which are almost the same, in spite of the difference between their densities. The values of the optical constants δ and β are presented in Table 1. For a precise determination of the parameters of the Cr and Fe layers, RXRR experiments with the wave energies close to the absorption edge of the above elements (5.989 KeV for Cr and 7.110 KeV for Fe) were performed at the "Phase" station of the Kurchatov Institute synchrotron source. In that experiment a Si(111) double crystal monochromator was used instead of Bartels 4xGe(220) one, the rest was not changed. All the RXRR curves were measured in θ-2θ mode.

The goal of the RXRR study was determine: a) the electromagnetic field distribution in the structure as a function of depth and b) the value of the refractive index for each layer. More detailed calculations of the reflectivity rocking curves were explained in [20,21].

In the X-ray wavelength range the refractive index n_i is a complex value, where the real part is a refraction coefficient and the imaginary part is an absorption one, and it can be expressed by the polarizability of the i-th layer χ_0^i, as shown in [22].

To obtain the distribution profile of the refractive index depending on depth, an inverse problem of the RXRR usually has to be solved. Based on the existing rocking curve, the solution helps to discern the microstructure model of the investigated sample. A possible way to get the solution is to apply search engine optimization methods using a chi-squared test χ^2. Herein all the data were analyzed simultaneously, because a separate calculation based on a few X-ray reflectivity experiments with different wavelengths can result in different models for the same sample with similar values of the χ^2 criteria. In the above approach a single model was used to refine the structure and single χ^2 criteria for the entire series of experiments calculated according to [23].

The simulations of RXRR curves were based on the Abeles matrix formalism [24]. To minimize χ^2 criteria, the Levenberg-Marquardt algorithm was utilized. The model of the periodic MLS was set as a number of layers with the following varied parameters: thickness, thickness of the transition area, density. The interfaces between the layers were described using the lamella approach. This approach implied the interface as a set of thin lamellas with smoothly varied δ and β parameters in the direction from the lower to the upper layer values. For the sake of keeping physical meaning, we chose the minimum thickness of the lamellas more than 0.2 nm (close to interplanar spacing). The total thickness of all the lamellas represented the size of the transitional area. A common approach to describe the interface roughness by entering Debye-Waller [25] or Nevot-Croce [26] factors was not explored, because their contribution was limited to an effective reduction of the electromagnetic waves amplitudes at the interfaces and did not account for refraction inside this area.

The definition of the MLS layer based on the split to sublayers with a constant value of the χ_0 and transitional part is shown in Fig. 2. The real and imaginary parts of the polarization ability dependence on depth is the subject of a certain distribution law, the cosine dependence is shown in Fig. 2.

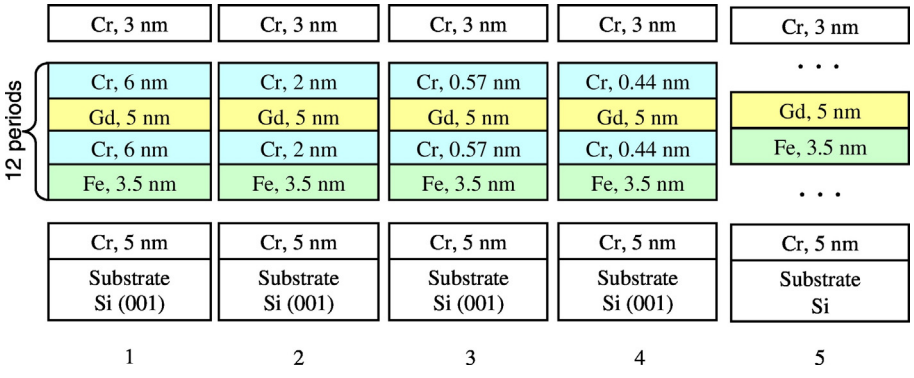

Fig. 1. Scheme of the samples. The sample No. are given at the bottom.