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SYNCHROTRON MÖSSBAUER REFLECTOMETRY OBSERVATION AND CELLULAR AUTOMATON SIMULATION OF DOMAIN FORMATION AND TRANSFORMATION IN ANTIFERROMAGNETICALLY COUPLED Fe/Cr MULTILAYERS

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Keywords: nuclear resonant scattering, reflectometry, magnetic domains, multilayers, cellular automaton

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1. Introduction

Antiferromagnetically (AF) coupled metallic multilayers (ML) have received much attention in recent years due to their relevance in fundamental science and magnetic recording technology alike. The performance of magnetoresistive devices is strongly affected by the ML domain structure [1]. Both plane-perpendicular and lateral magnetic structure of AF coupled metallic MLs can be efficiently studied by two closely related nuclear scattering techniques, viz. synchrotron Mössbauer reflectometry (SMR) [2, 3] and polarised neutron reflectometry (PNR) [4]. Here we present SMR studies of the magnetic-field-history-dependent formation and transformation of magnetic domains in strongly AF-coupled epitaxial MLs. One of the observed transformations will be described by a Monte Carlo simulation using a cellular automaton algorithm.

2. Experimental

An epitaxial MgO(001)/[^{57}Fe(2.6 nm)/Cr(1.3 m)]$_{20}$ ML was fabricated by MBE technique and was characterised reflection high-energy electron diffraction (RHEED), Rutherford backscattering (RBS), x-ray reflectometry, magneto-optical Kerr effect (MOKE), vibrating sample magnetometry (VSM), conversion electron Mössbauer spectroscopy (CEMS), conversion electron Mössbauer polarimetry (CEMP), specular PNR and specular SMR experiments. The room-temperature (RT) saturation field of the ML of fourfold in-plane anisotropy was found from the specular intensity of the SMR AF reflections to be H_S = 0.85 T and 1.05 T along the easy and hard axes, respectively. The easy-axis saturation field increased to H_S = 1.55 T at T = 15 K. A bulk-spin-flop (BSF) transition took place at RT when a magnetic field of 14 mT was applied along the easy axis in which the layer magnetisations actually lay [5, 6].

Diffuse SMR experiments were performed at the nuclear resonance beamline ID18 of the European Synchrotron Radiation Facility, Grenoble. The ML was placed in a liquid helium cryostat equipped with a superconducting solenoid and a variable temperature inset. The photons reflected from the ML were detected by an avalanche photo diode (APD).

3. Diffuse synchrotron Mössbauer reflectometry

The off-specular (diffuse) nuclear resonant reflectivity of synchrotron radiation (diffuse SMR) probes the in-plane component q_x of the scattering vector. In the kinematical approximation, the q_x-scan width at the AF Bragg peak (*i.e.*, at fixed q_z) is $\Delta q_x = 1/\xi$ where ξ is the in-plane correlation length ξ of the magnetisation, *i.e.*, the 'size' of the AF domains. A more accurate theory of diffuse SMR relating the diffuse scatter to the structural and/or magnetic autocorrelation function of the ML in terms of a distorted-wave approximation has been given recently [7].

4. Domain formation and ripening

Starting with a strongly AF-coupled ML in magnetic saturation and then gradually decreasing the field, two kinds of AF patch domains differing only in the sense of rotation of the magnetisation in their odd and even layers are spontaneously formed [8]. The patch domain formation is the consequence of the reduced stray field and the freedom of the AF-coupled regions in selecting the sense of rotation of their top-layer magnetisation during unsaturation [9]. On further decreasing the field and, thereby, increasing the domain-wall angle, the size of the domains is expected to spontaneously increase in order to decrease the domain-wall energy per unit area of the ML [10]. We observed this *domain ripening* with SMR. At RT, the native domain size of ξ = 370 nm did not change down to 200 mT while it spontaneously increased to ξ = 800 nm between 200 and 100 mT. No further increase of ξ was found down to remanence. The domain ripening was found to be an irreversible process and was followed by an apparent change in the shape of the diffuse SMR scattering peak. No ripening took place

at $T = 15$ K, probably a consequence of the temperature dependence of the coercivity [11]. However, the ripening was observed with SMR when the temperature was raised to RT. Domain ripening was not sensitive to the orientation of the external field relative to the magnetic easy axes.

5. Domain coarsening

A dramatic increase of ξ from 800 nm to at least 5 μm, *i.e.*, a *coarsening* of the AF domains was observed in the same multilayer [12] both with SMR and with PNR when it passed the BSF transition provided that the external easy-axis magnetic field was previously decreased from magnetic saturation to zero. In contrast to ripening, a domain-wall-energy-driven and coercivity-limited process, the explosion-like coarsening is driven by the Zeeman and the anisotropy energies and is not associated with any long-range domain-wall movement. Also coarsening was found to be irreversible as long as the applied field did not reach the saturation region.

6. Supersaturation memory effect

We observed an apparent *supersaturation memory effect* in the field history of the same Fe/Cr ML. At RT, we had to apply at least $H_{SS} = 1.30$ T in either easy or hard directions to erase the 'ripened' or 'coarsened' domain structure, *i.e.*, to convert the ML domains into their native size and shape. At $T = 15$ K, H_{SS} increased to a value as high as 3.60 T. A comparison with the behaviour of other Fe/Cr MLs suggests that this effect was the consequence of the presence of a small fraction of very strongly coupled regions in the first ML [13].

7. Cellular automaton simulation

Although the physical bases of these transformations are quite clear, so far no model has been able to quantitatively describe the observed details. In the following, we will present a simple model of the domain walls and of the domain-wall movement in strongly AF-coupled MLs. We will show that a cellular automaton simulation based on this model is able to describe all the observed details of domain transformations.

We describe the ML as consisting of AF domains, that are much bigger than the domain walls. The model magnetisation of the domains obeys a two-sublattice behaviour characterised by the opening angle 2φ calculated from the minimum condition for the bilinear layer-layer coupling and the Zeeman energy as $\varphi = \arccos H/H_s(\boldsymbol{r})$. We assume that the layer-layer coupling and, consequently, the saturation field $H_S(\mathbf{r})$ follow a certain (*e.g.*, Gaussian) distribution the expectation value and standard deviation of which are free parameters of the model. The domain-wall energy is supposed to be proportional to the square of the domain-wall angle with a coupling coefficient D having no lateral distribution. In addition, the coercivity H_c of the FM layers is considered constant for the whole ML but allowed to depend on the temperature T.

A micromagnetic simulation of the domain structure in a volume of the ML, which is large enough for calculating the autocorrelation function of the magnetisation with the accuracy necessary for determining the diffuse SMR scatter would include 10^{12}–10^{13} spins. This is technically not feasible and, therefore, the number of degrees of freedom has to be reduced considerably.

The Monte Carlo simulation starts with generating random values of $H_s(\boldsymbol{r})$ on a lattice of 'pixels' that will be the lattice points of a cellular automaton. One pixel is an area of the ML smaller than a domain (*i.e.*, the correlation length of the top-layer magnetisation) and bigger than a domain wall. One pixel consists of about 10^8 strongly coupled spins of the ML stack; the whole simulation includes 10^4–10^5 lattice points. The domain-wall energy of the ML is calculated as the sum of the next-neighbour pixel domain-wall energies with non-vanishing contribution only from pairs of opposite sense of rotation of the top-layer magnetisation. Should a pixel jump from one sense of rotation to the other, half of its full hysteresis loss, *i.e.*, $2H_cM \sin\varphi$ will be dissipated (M is the saturation magnetisation of one pixel).

The cellular automaton rule is to minimise the total energy of the lattice in monotonically changing external magnetic field or monotonically changing coercivity. The Hamiltonian contains a nearest-neighbour domain-wall energy term as well as the dissipative penalty term describing the hysteresis loss. Further terms of the Hamiltonian, *i.e.*, the Zeeman energy and the bilinear layer-layer interaction of random lateral distribution are replaced by the logical condition $\varphi = 0$ for $H \geq H_s$ and $\varphi \neq 0$ for $H < H_s$. Indeed, both terms are independent of the sense of rotation.

The movies of the domain dynamics with varying H or T (*i.e.*, H_c) to be shown in the presentation consist of pictures. Subsequent pictures of the calculation always differ from each other only by the sense of rotation of a single pixel (the saturation state being considered to have a third, 'neutral' sense of rotation). On gradually changing H or H_c, a pixel will change its sense of rotation if the new state, taken into account the domain-wall energy and the hysteresis loss, will be energetically more favourable. Thus, the simulation depends from D and H_c only through their ratio D/H_c. The simulation of domain ripening reproduces the observed, relatively sudden transition. This is attributed to the fact that the pixel model properly accounts for the local character of the domain-wall interaction. Therefore ripening is seen in the movies as a smoothing out of the domain walls and vanishing of small enclosures of the opposite sense of rotation. Other H-dependent simulations fairly well describe the supersaturation behaviour in case of a broad distribution of $H_s(\mathbf{r})$. Starting from the native state, and decreasing H_c (*i.e.*, increasing T) [11] the temperature-induced ripening is well simulated.

8. Conclusion

In conclusion, we have shown that the domain structure of a ML of strong AF coupling depends on the history of the external magnetic field and of the coercivity of the

ferromagnetic layers (*i.e.*, of the temperature). This results in a great variety of domain formation and transformation processes. The cellular automaton algorithm based on the rough 'pixel model' of the domains depending only on a very small number of free parameters is able to properly reproduce the main features of domain ripening and domain memory supersaturation in AF-coupled MLs.

Acknowledgements: The authors thank F. Iglói, Yu.N. Khaidukov, L. Lovász, Yu.V. Nikitenko, A.V. Petrenko, V.V. Proglyado and Gy. Szabó for stimulating discussions. Provision of synchrotron radiation by the European Synchrotron Radiation Facility as well as financial support by the Hungarian National Fund (OTKA) and by the European Community under the respective contract Nos. T047094 and STREP NMP4-CT-2003-001516 (DYNASYNC) is gratefully acknowledged.

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