IO-10 **Wed. 18. 06., 15^{00}-15^{20}**

Relations between structural and magnetic properties in metallic ultrathin multilayers

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Keywords: magnetic multilayers, magnetic anisotropy, interlayer coupling, synchrotron radiation

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Ultrathin magnetic multilayers attract scientific interest as excellent objects for studying nanomagnetism. They are of high technological relevance as potential materials for novel practical applications as well. Due to dimension confinement and substantial amount of atoms forming surfaces or interfaces such structures exhibit novel properties not encountered in bulk materials.

Investigations of structural features and manipulation at the atomic scale in correlation with observed magnetic properties allow on the one hand to understand physical phenomena and on the other hand to design and fabricate nanosystems with desired attributes. However, structural studies of ultrathin film are a challenge. Due weak signal from low amount of investigated material standard techniques cannot probe these properties reliably. Techniques exploiting X-ray (e.g. X-ray reflectivity, XRR) or synchrotron radiation are only methods enabling to gain an insight into atomic ordering (e.g. X-ray absorption near edge structure, XANES) and magnetic properties (e.g. X-ray magnetic circular dichroism, XMCD). Particularly, element sensitive synchrotron techniques are very powerful for investigations of low amounts of material.

In this work we focus attention mainly at the structural and magnetic properties of ultrathin Co/Mo multilayered system. Undertaken research has been inspired by the interface influence observed in our earlier studies of magnetic nanodots with perpendicular magnetization [1, 2] induced in ultrathin Co layer deposited on a patterned buffer in a form of Au islands self-assembled on the Mo buffer surface [3]. The Co/Mo system is particularly interested as the crystallographic difference of Co (hcp) and Mo (bcc) and lattice parameters allow for intentional modifications of magnetic layer structure and interfaces which substantially affect magnetic properties. It is worth also to mention that cobalt / nonmagnetic early transition metal systems in general have been studied to less extent, so far, and their properties are not recognized well, yet.

Investigated Co/Mo sandwiches, containing a single Co layer, or multilayers with higher repetition number of bilayers were grown on sapphire substrates in a molecular beam epitaxy (MBE) system. Dependently on measurements to be carried out thickness of the component layers was uniform or they were grown as wedges with use of a linear shutter moving during material deposition. The growth mode was monitored in-situ by reflection high electron energy diffraction (RHEED). This technique allows to determine a growth type (amorphous, polycrystalline or monocrystalline) and the symmetry of crystallographic structure and directions in monocrystalline layers. Moreover, it enables to estimate the strains induced by the lattice mismatch at the interfaces. Magnetic properties were measured exploiting magnetooptical Kerr effect (MOKE). Both polar (PMOKE) and longitudinal (LMOKE) configurations of the setup were used. Local probing of the magnetization is a very important advantage of this technique. In combination with wedge type structures it allows in an easy way to study magnetic properties in a function of a component layer thickness. Particularly, the sandwiches containing two wedge-like layer with orthogonally oriented thickness gradients enable to obtain two dimensional diagrams of magnetization processes and related parameters such as e.g. coercivity, effective anisotropy field or remnant magnetization. X-ray reflectivity was applied to study layered structure of sandwiches and quality of interfaces. Numerical fitting of obtained experimental spectra allows to confirm assumed thicknesses of the component layers and to determine roughness (or alloying extent) of the interfaces. Synchrotron radiation measurements were carried out in European Synchrotron Radiation Facility (ESRF) in Grenoble at the beamline ID12. Crystalline structure of grown Co ultrathin layers was inferred from XANES measurements at Co K edge. Magnetic moments induced at Mo atoms due to proximity with adjacent Co layers were determined from XMCD investigations performed at Mo $L_{2,3}$ edges in the magnetic field as high as 0.9 T.

Magnetization orientation in ultrathin films is governed by the effective magnetic anisotropy being an interplay of several components. Shape (dipolar) anisotropy results from the interaction of the magnetization with magnetic field generated by magnetic charges appearing in the perpendicularly magnetized films at the interfaces due to broken symmetry. This component always forces magnetization alignment in the sample plane. Remaining constituents may orient magnetization both in perpendicular direction to the sample plane or in the sample plane. Magnetocrystalline anisotropy is related to minimum energy of magnetization along low index crystallographic directions. Magnetoelastic anisotropy is associated with a lattice deformation. In thin film structures such deformation occurs mainly due to lattice misfit at the interfaces. Particularly, in the Co films with hexagonal (hcp) structure the anisotropy easy axis is parallel to c-axis. Under tensile strains magnetization rotates toward perpendicular direction to the strain axis. Thus for Co hcp layer with c-axis perpendicular to the film plane in-plane tensile strains enhance its perpendicular magnetic

anisotropy. A spin-orbit coupling is responsible for these two kinds of anisotropy. Finally, an interface anisotropy is another component that occurs in thin film structures. It originates from the broken symmetry in the surrounding atoms. The electronic structure of atoms forming the surfaces or interfaces may be modified due to non-saturated dangling bonds or electron hybridization with adjacent atoms of a different type, respectively.

As a result of anisotropy thickness dependence a spin reorientation transition (SRT) is a commonly observed phenomenon in the layered structures containing an ultrathin Co layer. Due to mentioned above effective anisotropy the magnetization of thin Co layer sandwiched between non-magnetic covers is oriented perpendicularly to the plane. At a critical thickness of the Co layer, d_{SRT}, it rotates towards in-plane alignment. The value of d_{SRT} depends on the covers type. One of the highest values are obtained for Au and Pt covers being equal to 1.8 and 2.2 nm, respectively [4, 5]. Such behavior is well described by a model of uniaxial anisotropy taking into consideration contributions from Zeeman energy and from effective anisotropy described by the coefficient K_{1eff}.

On the contrary to noble metal covers in Mo/Co/Mo sandwiches the SRT is not observed. It means that the Co layer is magnetized in the sample plane in the whole range of its thickness. The same magnetization alignment is found in the Co/Mo structures containing two or three Co layers. Additionally, from LMOKE investigations a strong magnetic anisotropy in the sample plane is inferred. The angular dependence of remnant magnetization, m_R and coercivity, H_C clearly prove a two-fold symmetry. This in-plane anisotropy is the common feature of structures consisting of one, two and three Co layers. The Co/Mo multilayers exhibit also very well distinguished antiparallel (antiferromagnetic) coupling of magnetization of the Co component layers. Such type of coupling is observed for the Mo spacer in the thickness range between 3 and 6 atomic layers (AL) and for the Co layer thickness of the order of several nanometers. The LMOKE magnetization curves consist of characteristic rectangular sub-loops of number corresponding to the number of Co layers in the studied sample. With farther increase of the Mo spacer thickness the interlayer coupling adopts a parallel order (ferromagnetic character). For that type of alignment the hysteresis loop exhibits a rectangular shape.

To explain magnetic behavior described above we studied structural properties of the multilayers in details.

Streak-like RHEED pattern proved the epitaxial character of Co/Mo multilayers at every stage of the growth process. However, non-uniform spotty structure of the streaks suggests roughness occurrence at the interfaces. To confirm assumed nominal structure of the studied multilayers the XRR measurements were carried out for uniform multilayers displaying mentioned interlayer magnetic couplings. The reflection spectra consist of expected Bragg peaks and Kiessig fringes. The parameters describing the structure were determined from the fitting procedure of calculated spectra to that obtained in the experimental way. Achieved thicknesses correspond well with assumed ones and were equal to 4 nm for Co layers and 0.8 and 1.8 nm for Mo spacers in the samples exhibiting antiparallel and parallel magnetic coupling, respectively. Interface roughness for those samples was equal to 0.3 and 0.5 nm. It correlates with spotty character of RHEED streaks.

In-plane magnetization alignment of the Co layers in the whole thickness range in Co/Mo multilayers indicates that sum of crystallinity, interface and strain contributions to magnetic anisotropy does not outweigh the shape influence. Thickness dependent crystalline structure of the Co layers was clearly inferred form XANES measurements. For this purpose we studied two Co/Mo multilayers with the same Mo spacer thickness, equal to 1.8 nm and different Co layer thicknesses of 0.9 nm and 4.1 nm. The absorption profiles at the Co K edge for these two samples differ substantially each other particularly in a shoulder shape around 7715 eV and the peaks appearing above 7720 eV. Comparison of experimentally achieved profiles from our samples with the profiles measured for intentionally grown fcc and hcp Co layers, simulated also numerically [6], evidently shows that the crystalline structure of the Co layer 0.9 nm thick is of fcc type whereas for thicker layer hcp stacking dominates. Thus one can draw a conclusion that for Mo buffer or spacer the Co layer grows in the initial stage in fcc structure. Such growth mode explains well magnetic properties of the Co layer deposited on Mo buffer. Magnetic anisotropy forcing perpendicular magnetization is much weaker for fcc structure. The structural evolution from fcc to hcp is also reflected well in the Co layer thickness dependence of effective anisotropy field H_{1eff} directly related to effective anisotropy characterized by the coefficient K_{1eff}.

A mechanism responsible for the observed interlayer coupling remains still undiscovered. Ruderman–Kittel–Kasuya–Yosida (RKKY) oscillating interaction mediated by conduction electrons is one of the candidates. A model of quantum well (QW) assuming spin-selective electron scattering at the interfaces can be also taken into account. In another approach the magnetic poles appearing at the interfaces due to their roughness may also couple the magnetic layers through magnetostatic interactions. To justify a nature of the interlayer coupling XMCD measurements probing magnetic moments at Mo atoms of the spacer were performed. The absorption was investigated at the Mo $L_{2,3}$ edge. Two studied multilayers differed in Mo spacer thicknesses equal to 0.8 nm and 1.8 whereas the Co layer thickness of ca. 4 nm was the same in both samples. Additionally, a reference film grown as a uniform alloy $Co_{96}Mo_4$ was also studied. In the alloyed film induced magnetic moment at Mo site was expected to be strongest. The measurements reveal this moment in the alloy sample as high as -0.21 μ_B. A moment weaker by almost an order of magnitude was found in the layered sample with thinner Mo spacer. In these two samples the induced moment was coupled antiparallel to the moment of adjacent Co atoms. The XMCD signal is below the detection limit for the layered sample with the thickest Mo spacer. One of possible interpretations of relatively large difference in detected induced moments at Mo site in alloy and the spacer is that the moments are aligned antiparallel in the

consecutive individual atomic layers of the Mo spacer. Such alignment of moments induced in W spacer has been found in Fe/W system [7]**.** For this purpose the measurements of X-ray resonant magnetic scattering (XRMS) measurements, i.e. a combination of X-ray circular magnetic dichroism (XMCD) with anomalous X-ray reflectivity (XRR) are planned in the next step of research.

Acknowledgments: This work is supported by the research funds from the National Science Centre in Poland under the projects: DEC-2011/03/N/ST3/02662 and DEC-2012/06/M/ST3/00475.

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