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DETERMINATION OF THE DIRECTION OF LAYER MAGNETIZATION USING ELLIPTICALLY POLARIZED RESONANT PHOTONS

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Introduction

Modern industries, in particular the IT and automotive industries use magnetic multilayer structures in a number of areas. The applications are principally based on the Giant Magnetoresistance (GMR) phenomenon. The relative alignment of the magnetization of ferromagnetic layers in the multilayer are manifested in a significant change of the resistance of the multilayer (by several percents, Baibich, 1988), and, since this relative alignment is changed by an external magnetic field it is an extremely sensitive way of magnetic field sensing. The antiferromagnetically coupled periodic Fe/Cr multilayer was the first example of an artificial antiferromagnetic (AF) layer structure. Later other metallic systems were also found AF-coupled. The GMR allows for sensing the value of the magnetic bit irrespective of the rotational speed of the hard disk, which opened giant perspectives in increasing the areal information density. The feasibility of the GMR read head was demonstrated in 1994. In 1997, the first devices appeared on the market. At the Millennium, each sold hard disk had read heads of GMR type. The scientific and technological importance of GMR was so immense, that the 2007 Nobel Prize in Physics was given to A. Fert and P. Grünberg for the discovery of GMR. The development has continued ever since. Researchers and engineers found new solutions to increase the areal density and the reading speed. Read heads based on the tunelling magnetoresistance (TMR) allow for even smaller bits and higher areal density than GMR-based ones, since their specific change in the magnetoresistance is bigger. In recent years, perpendicular magnetic recording (PMR) appears to be in the forefront of development, since the data density that is approaching its theoretical limit can be increased by about one further order of magnitude.

The above historical survey demonstrates that the direction (alignment and sign) of the layer magnetization is an important piece of information both from basic science and from practical application points of view. It conveys information about the coupling of the magnetic layers, the direction of the magnetic domains, and about of the magnetic anisotropy.

The direction of the local magnetization determines the direction of the hyperfine field. In case of ^{57}Fe Mössbauer resonant nuclei and metallic iron they are exactly anti-parallel. Hyperfine fields can be measured by Mössbauer spectroscopy; consequently the

Mössbauer-effect-based spectroscopic methods are appropriate for the determination of the alignment and sign of magnetization of (possibly buried) layers. In the traditional Mössbauer spectroscopy the transmitted radiation is detected, which is impossible in case of multilayers on perforce thick substrates. For thin layer Mössbauer studies can be performed by detecting the conversion electrons which also originate from the resonance process (conversion electron Mössbauer spectroscopy, CEMS). However, prior to this work, neither experimental conditions, nor theoretical foundation of CEMS polarimetry, i.e. CEMS using polarized source and sample, were available. The present work is a study of this important topic.

Aims of the present work

The present theses deal with methodological, data evaluation and applications aspects of CEMS polarimetry. The aims of the present work are:

- To establish polarimetry in conversion electron and synchrotron Mössbauer spectroscopies, by developing and fabricating the necessary devices for those,
- To develop methods to evaluate Mössbauer polarimetric spectra, and
- To apply CEMS polarimetry to study the variation of the layer magnetizations in antiferromagnetically coupled structures.

Applied methods

The determination of the direction of layer magnetization is not a simple task in the case of antiferromagnetic structures. The traditional magnetometric methods (e.g. vibration sample magnetometry (VSM), SQUID magnetometry, magneto-optical Kerr-effect magnetometry (MOKE)) integrate over some depth and have no or limited resolution for the individual layer magnetizations. In Mössbauer polarimetry, improved by us, the resolution is suitable to resolve individual layer magnetizations. However, the realization of polarimetry in thin-film-sensitive geometry is not quite obvious. The Mössbauer polarimetry, herewith by which I name all polarized-source Mössbauer spectroscopic studies, is sensitive to the direction of the hyperfine magnetic field. Combining CEMS and polarimetry, magnetic multilayer samples can be studied. In CEMS the thick substrate is not a drawback, since it is a back-scattering method. The CEMS polarimetry further combined with the isotope-marker technique makes the direction of the magnetization of a single atomic layer possible to determine.

Synchrotron Mössbauer spectroscopy opens further perspective of the method, the synchrotron beam being focused, allowing for lateral resolution not present in traditional Mössbauer spectroscopy. For the experiments to follow we needed multilayer samples of adequate quality. The samples were prepared by molecular beam epitaxy (MBE), earlier in the laboratory of IKS, our Belgian partner Institute, later in Hungary's first, and presently only MBE system, which has been installed in our Institute.

Theses

I. I have demonstrated the linear Mössbauer polarimetry in transmission geometry using a ^{57}Co(α-Fe) source. CEMS was never used before for external field magnetization studies. I have shown that the electron deflecting effect of the external field upon the conversion electrons is negligible, and does not reduce the efficiency of the CEMS detector. I have designed and built the first ever (^{57}Co(α-Fe) source) *linear* CEMS polarimeter, and I experimentally demonstrated, that polarimetry can be performed in CEMS [1].

II. I have demonstrated the elliptical Mössbauer polarimetry using ^{57}Co(α-Fe) source in transmission geometry. In order to demonstrate that the method is applicable to CEMS, I have designed and built the first ever (^{57}Co(α-Fe) source using) *elliptical* CEMS polarimeter [2].

a) We designed and built a unique sample magnetizer which is suitable for performing full-magnetic-loop experiments and which consists of six permanent magnets, the net field of which is parallel to the optical axis [2].

b) We designed and built a unique CEMS detector for low angle and perpendicular incidence of γ–photons [2] for elliptical Mössbauer polarimetry. The detector was successfully used by our Polish partners in their circular polarimetric experiments using a ^{57}FeSi filter [3].

III. I have developed a new polarimetric method in synchrotron Mössbauer spectroscopy, in which the originally linear polarization of the synchrotron radiation is converted into (almost) circular polarization using a Zeeman-split nuclear resonant filter (a longitudinally magnetized ultrathin ^{57}Fe foil) [4].

a) By forward scattering experiments I have demonstrated the applicability of the synchrotron Mössbauer polarimetry [4].

b) By grazing incidence experiments I have shown that synchrotron Mössbauer polarimetry is applicable to determine the (alignment and) the *sign* of the layer magnetization in thin films.

IV. I have shown, that an elliptical Mössbauer polarimetric spectrum – in the case of thin source and thin absorber – is a linear combination of four standard subspectra. From the four (three independent) coefficients I have determined the polar angle of the hyperfine magnetic field, and also the difference of the two azimuth angles in the source and in the absorber, respectively [5].

V. Applying the evaluation method of IV,

a) I have shown experimentally, that the evaluation method is correct for the perpendicular-incidence spectra and that the reason for inaccuracies in the case of the low-angle incidence is due to multiple scattering, i.e. the thickness-effect [2].

b) Based on Szilárd Sajti's theory I have shown how to account for thickness effects in low-angle-incidence polarimetric spectra. We have developed a method to evaluate integral CEMS measurements performed with polarized sample and source and gave – for the first time in literature – the correct relative intensities for thick (or tilted) samples [6].

VI. As an application of all the above, I have examined the behavior of the layer magnetizations in ^{57}Fe/Cr multilayer antiferromagnets.

a) I have demonstrated the bulk spin flop phenomenon by linear polarimetry [1]. This is a unique result in laboratory conditions although the bulk spin flop was demonstrated earlier by polarized neutron reflectometry (PNR), and synchrotron Mössbauer reflectometry (SMR).

b) Using linear Mössbauer polarimetry I studied the alignment of layer magnetizations with the external field along the easy axis of Fe. The results could be satisfactorily described by a model using the literature values of the specific magnetization and bulk anisotropy coefficient of iron, as well as the literature value of the layer-layer exchange coupling constant in Fe/Cr, taking account finite-size effects.

c) Saturating and relaxing the Fe/Cr AF multilayer in the exact magnetic hard direction, four kinds of domains form which are mutually perpendicular to each other (Rührig state). I was able to reproduce this fully compensated state in the linear CEMS polarimeter. Using diffuse SMR, I found, that the field-parallel domains are enlarged, however field-perpedicular domains remain small. On the same sample, consequent to the same magnetization history, the change of the width of the diffuse scatter peak was not observable by PNR. In my interpretation, the magnetization structure of the multilayer forms a surface spin flop state, the top and bottom layers being mirror images of each other. Consequently, for the high penetration depth neutrons the magnetizations are averaged, and no effect is observed, while for the 0,086 nm wavelength synchrotron radiation of much lower penetration depth the top stack of layers contribute much more to the diffuse signal than the bottom stack and the effect is visible. The difference of the two results is an indirect experimental evidence of the surface spin flop state.

d) By linear Mössbauer polarimetry, I have shown the spin rotation during relaxing the field from saturation by a field oriented by some degrees off the exact hard direction. The results and simulations on this system show, that, upon relaxing the field, the layer magnetizations evolve by surface spin flop into that easy direction, which became preferred by the misorientation.

e) I have followed the evolution of the magnetization in an asymmetric MgO(001)/^{nat}Fe/Cr/^{57}Fe epitaxial trilayer upon a magnetic cycle. I found an uniaxial spin flop at 20 mT. These results are unique in laboratory conditions.

VII. I have studied the Gd(7nm)/^{57}Fe(7nm)Gd(7nm) structure by elliptical synchrotron Mössbauer polarimetry, below and above the 130 K compensation temperature of the system. As expected, above the compensation temperature, the ^{57}Fe spectrum can be described by a field-parallel homogeneous Fe layer magnetization. Below the compensation temperature, where the two Gd layers' magnetization exceeds the Fe magnetization, however, an inhomogeneous magnetization of broad angular distribution was observed instead of the expected homogeneous Fe layer magnetization. This was qualitatively interpreted using a magnetic exchange spring model.

Conclusions

Mössbauer polarimetry is a well suited method in determining the direction of the magnetization. It can be used in conversion electron Mössbauer spectroscopy, an appropriate method to study ^{57}Fe containing multilayer structures. In layer antiferromagnets phenomena, like bulk spin flop could be detected using Mössbauer polarimetry, which earlier were only possible by PNR and SMR at large facilities. The synchrotron version of Mössbauer polarimetry is particularly appropriate to study thin film magnetization phenomena in a wide temperature and magnetic field range.

List of publications

Publications directly connected to theses

[1] F. Tanczikó, L. Deák, D.L. Nagy, L. Bottyán; „Conversion electron Mössbauer spectroscopy with a linearly polarized source"; Nuclear Instruments Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **226** 461–468, (2004)

[2] F. Tanczikó, Sz. Sajti, L. Deák, D.G. Merkel, G. Endrőczi, D.L. Nagy, L. Bottyán, W. Olszewski, K. Szymanski; „Electron proportional gas counter for linear and elliptical Mössbauer polarimetry"; Review of Scientific Instruments **81** 023302 (2010)

[3] W. Olszewski, K. Szymański, D. Satuła, L. Dobrzyński, L. Bottyán, F. Tanczikó; „Magnetic texture determination by Conversion Electron Mössbauer Spectroscopy with circularly polarized beam"; Nuclear Instruments Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **266** 3319–3324 (2008)

[4] F. Tanczikó, L. Bottyán, A.I. Chumakov, B. Croonenborghs, L. Deák, J. Korecki, J. Meersschaut, D.G. Merkel, D.L. Nagy, N. Planckert, R. Rüffer, H.D. Rüter, Sz. Sajti, C. Strohm, E. Szilágyi, K. Szymański, Á. Tunyogi, V. Vanhoof; „Sign Determination of the Hyperfine Field by Elliptically Polarized Nuclear Resonant Synchrotron Radiation"; Nuclear Instruments Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, before submission

[5] F. Tanczikó, L. Bottyán, L. Deák, D.G. Merkel, D.L. Nagy; „Sign Determination of the Hyperfine Field by Elliptically Polarized Mössbauer Source"; Hyperfine Interactions **188** 79–84 (2009)

[6] Sz. Sajti, F. Tanczikó, L. Deák, D.L. Nagy and L. Bottyán; „Angular dependence, blackness and polarization effects in integral conversion electron Mössbauer spectroscopy"; Physical Review B, before submission; arXiv:0912.1539v1

Further publication on the present topics

F. Tanczikó, L. Deák, D.L. Nagy, L. Bottyán; „Thin Film Studies with Magnetic-Field CEMS Detector: Mössbauer Polarimetry with Linearly Polarized Source"; *Condensed Matter Studies by Nuclear Methods* Proc. XXXVIII. Zakopane School of Physics, Zakopane, pages 175–182. Jagellonian University Kraków, (2003)