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Review of doctoral dissertation entitled
Tailoring the magnetic anisotropy of antiferromagnetic thin films in epitaxial multilayer systems and ferromagnetic/antiferromagnetic nanostructures
by Hashim Nayyef

Doctoral dissertation entitled *Tailoring the magnetic anisotropy of antiferromagnetic thin films in epitaxial multilayer systems and ferromagnetic/antiferromagnetic nanostructures* prepared by Hashim Nayyef is devoted to the interaction between antiferromagnetic (AFM) and ferromagnetic (FM) thin films. This type of heterostructures (AFM/FM) belong to a group of materials revealing interesting physical properties including exchange bias which very soon can find vast applications. Specifically, they can be used as building blocks of spintronic devices used in data storage and sensing magnetic field.

In his thesis Hashim Nayyef presents results of investigation of fundamental magnetic properties of NiO and CoO antiferromagnets prepared on Fe layer without and with nonmagnetic Au spacer. The studied heterostructures were fabricated using molecular beam epitaxy method. In his research Hashim Nayyef uses surface sensitive techniques including low energy electron diffraction (LEED), magneto-optic Kerr effect (MOKE), X-ray magnetic circular dichroism (XMCD), X-ray magnetic linear dichroism (XMLD) and associated photoelectron emission microscopy (PEEM) technics: XMCDPEEM and XMLDPEEM. The reported experiments were performed in part in laboratory systems and in part at Solaris synchrotron facility in Krakow.

Doctoral dissertation of Hashim Nayyef has been completed at Faculty of Physics and Applied Computer Science at AGH University of Krakow under supervision of Dr. hab. Eng. Michał Ślęzak. The dissertation consists of nine chapters, it is written in English and counts 103 pages.

The first chapter is an introduction in which the Author explains the most important aspects of the dissertation. He emphasizes the importance of antiferromagnetic materials showing, for example, the disadvantages of ferromagnets in comparison to AFM/FM heterostructures.

Chapter two is devoted to introduction to magnetism of thin films. In particular, the Author presents basic information on magnetic anisotropy, its origin in bulk materials and nanostructures including magneto-crystalline, shape and surface anisotropies. He also discusses magnetic properties of Fe films grown on W(110) substrate, including thickness and temperature induced spin reorientation transition. The system is important from the point of view of the dissertation.
A relatively large part of the second chapter is devoted to antiferromagnets and FM/AFM heterostructures. The Author introduces fundamental properties of this class of magnetic materials and discuss most important phenomena: exchange bias and interfacial coupling between FM and AFM.
A comment to this chapter:
The Author claims that ... *at the critical thickness, it takes no energy to switch the magnetic moments from [001] to [1-10] and vice versa.* (p. 20). Basically, it means that the energies of both states are equal. However, in order to switch between both states one needs to overcome energy barrier which is between both directions. Such barrier is even shown by the Author in Fig. 2.3 b.

In chapter three Hashim Nayyef presents research methodology. Beside basic information on molecular beam epitaxy (MBE) he introduces an ultrahigh vacuum system in which samples used in the performed experiments were prepared. In the next subsections experimental methods are discussed including LEED, MOKE, XMCD and XMLD.
Comment to the third chapter:
In the following chapters the Author uses $R_{L,2}$ and $R_{L,3}$ parameters as a measure of *the XMLD magnitude* which is defined as a ratio of the corresponding peaks in the XAS spectra. In my opinion it would be good to have more in depth introduction of the physics behind these parameters and explanation why they are measure of the XMLD magnitude.
And smaller remarks:
In Fig. 3.1 a deposition source is denoted as MBE.
In Fig. 3.8 caption the Author states: *The particular substrates are denoted by quantum numbers…*
What does Author mean by substrates?

The next three chapters present results of measurements together with discussion. They include three studied layered systems: NiO(111)/Fe(110), CoO(111)/Fe(110) and CoO(111)/Au(111)/Fe(110).

In Chapter 4 the Author reports on the investigation of the NiO(111) layer grown on the Fe(110) films. In a short introductory part Hashim Nayyef informs that the studied system belongs to a group of AFM/FM heterostructures in which magnetism is controlled by tuning magnetic anisotropy of the ferromagnetic film. The Author prepared samples with the Fe thicknesses assuring the easy axis of magnetization aligned either along the [1-10] or [001] direction. In addition, he made series of samples with Fe thicknesses close to the thickness at which spin reorientation transition occurs where magnetic anisotropy is weak.

The obtained spectroscopic (XAS, XMLD) and microscopic (XMCDPEEM, XMLDPEEM) results indicate that the magnetic anisotropy of ferromagnetic iron film determines the easy axis of antiferromagnetic layer. Although NiO has threefold symmetry it reveals twofold in-plane magnetic anisotropy like in the Fe film below. It is also clearly shown that that thickness-induced SRT occurring in the Fe film drives 90° in-plane rotation of magnetic moments in NiO layer.

There are two more interesting achievements reported in this chapter. First is the observation of rotation of the easy axis of AFM layer coupled to FM film using external magnetic fields as low as 500 Oe. This very low magnetic field is of orders of magnitude lower than necessary for switching easy axis in isolated AFM layers. The other very important result is temperature-induced switching of the in-plane easy axis in NiO. It is driven by the SRT occurring in the Fe layer which has a thickness close to the critical SRT thickness and SRT is caused by heating or cooling the film.

Questions regarding chapter four:

1. The presented LEED patterns for 50 Å and 150 Å Fe without and with NiO layer on top are supposed to show that there are no structural (crystallographic) differences between the samples. However, there are various distances between the corresponding Fe diffraction spots recorded for 50 Å and 150 Å in both crystallographic directions (Fig. 4.2) while the intensity profiles shown in Fig. 4.3 show no differences in a distance between the corresponding maxima. What is the reason for this discrepancy?

2. Why a shape/character of the temperature dependence of $R_{L,2}$ shown in Fig. 4.11b is different from the curve presented in the lower part of Fig. 4.10? Instead of steep increase of the $R_{L,2}$ there is a jump to lower values?

3. What does a small, broad maximum between main peaks mean (Fig. 4.3 right, lower plot)?

Chapter five considers magnetic properties of the CoO(111)/Fe(110) layered structure. After a short introduction describing main topic of that part of dissertation and a paragraph devoted to sample preparation conditions the Author reports on the obtained results. Specifically, the studies of exchange bias and spin orientation of the antiferromagnetic layer at the interface with the Fe film are presented. Similarly like in the previous chapter the experiments have been performed for the two characteristic Fe film thicknesses: below SRT (50 Å) and above SRT (200 Å). According to the MOKE measurements the investigated system reveals exchange bias which is determined by the magnetism of Fe film above the Néel temperature of CoO. It appears that the direction of magnetic moments in the Fe layer (which depends on the Fe thickness) aligns magnetic moments in the neighboring CoO layer even above the Néel temperature. In this way it is shown that the magnetic anisotropy of AFM layer can be controlled by the magnetization of the FM layer below.

Comment to chapter five:

The author states that: *The XAS spectra for the two Fe thicknesses show significant differences after the sample has been cooled down…, see Fig. 5.1 (d).* In my opinion there is no significant difference between both curves at least in Figure 5.1 (d).

Chapter six is devoted to the CoO(111)/Au(111)/Fe(110) multilayers. The Author investigates in detail the influence of nonmagnetic spacer introduced between AFM and FM layers on magnetic anisotropy of ferromagnetic film and the resulting magnetic order of antiferromagnet.
In a short Introduction Hashim Nayyef presents motivation for studying AFM/FM system with additional Au layer. In the sample preparation part he includes a sketch of the orthogonally wedged sample which shows an overview of variety of the studied subsystems and on the other hand on amount of data and information which could be obtained as an output from the performed experiments. He also presents the LEED patterns obtained during subsequent steps of deposition showing epitaxial growth of all three sublayers.
The first figure presented in the results subsection gives an example of variety of data that can be obtained from such double-wedged sample. The MOKE microscope image together with the corresponding hysteresis loops taken at different regions of the image and with the external magnetic field along two orthogonal directions give a taste of diversity and complexity of the obtained results.
In the presented analysis of such rich set of data the Author considers six characteristic regions identified in the MOKE microscope image and compares corresponding results. The detailed discussion leads to numerous conclusions. Among the most important is the use of changes in the magnetic anisotropy of the ferromagnetic layer caused by a direct contact with a gold film in order to tune magnetic properties of antiferromagnetic layer. The interaction between FM and AFM layers depends on a thickness of the gold film causing switching of the easy axis from [1-10] to the [001] direction for enough thin Au films. Such rotation is caused by the freezing effect of interfacial CoO spins and the resulting introduction of additional magnetic anisotropy promoting [001] direction as the easy axis. The situation changes with increasing thickness of the gold film which decreases exchange interaction between AFM and FM layers.
The other interesting result is a strong influence of exchange bias on magnetic anisotropy of ferromagnetic film resulting in the rotation of the easy axis of Fe to the [001] direction after field cooling in the case of Fe film's thicknesses for which [1-10] easy axis is expected. This unexpected switching of magnetization direction has been explained by the presence of exchange bias which is a source of additional magnetic anisotropy driving the observed rotation. The phenomenon occurs in the CoO/Au/Fe multilayer system with the Au spacer thicknesses up to 15 Å. Above that value the interaction between AFM and FM becomes weaker leading to significant decrease of exchange bias and as a consequence the rotation of the Fe magnetization back to the [1-10] direction.
In this way the Author introduced another way, by interface engineering, of tuning magnetic anisotropy in the AFM/FM multilayers.

The entire doctoral dissertation is summarized in chapter seven. The Author, in a few sentences, presents each of the three investigated multilayer systems explaining how to control the direction of magnetization in an antiferromagnetic material.
The doctoral thesis is finished with the Reference section containing 152 citations and the list of nine author's publications.

The layout of the thesis is correct and the chapters form a logical sequence. Chapters five and six presenting experimental results are concluded with short summaries. For some reason chapter four does not have such summary.
The dissertation is written in a very condense way, like article, although it is not in a form of collection of the published papers. It presents results of experiments performed for several different AFM/FM systems prepared using the wedge layers. Therefore, in my opinion, taking into account the diversity of the obtained results, the dissertation could be more descriptive.

Below is a list of technical remarks.
1. Different spellings of the same word, e.g. *magneto-crystalline* vs. *magnetocrystalline.*
2. No verb in a sentence: *... where the magnetization transitions from an out-of-plane orientation at low temperatures to the in-plane of the film at around 300 K.*
3. Unfortunate phrases: *In Fig, 2.5, and at 300 K, Fruchart et al. found that...* (p. 23).
4. In some cases abbreviations appear before their explanations, e.g. UHV.
5. There is no explanation of symbols used in Fig. 3.3 in the figure caption.
6. In several cases Figure captions appear in part or in full on the next page (e.g. Fig. 4.2, Fig. 4.5, Fig. 5.3, Fig. 6.4).
7. Numbers on the coordinate axes are sometimes very small (e.g. Fig. 4.3) or not correctly displayed (e.g. Fig. 4.8).
8. There seems to be no consistency in open and closed symbols in Fig. 4.5b.
9. Below the Fig. 4.9 caption a new paragraph starts which looks like a repetition of the Fig. 4.9 caption.
10. Two phrases: *can be observed* and *can be seen* in one sentence, p. 68.
11. Imprecise phrase: *the vector of incoming linearly polarized X-ray beam was parallel to the Fe[1-10] in-plane direction*, p. 69.
12. Gray loops in Fig. 5.3 are barely visible.
13. Additional information e.g. legends in Fig. 5.3e-h are very small; it's hard to read it.
14. In the caption of Fig. 5.3 Fe thickness is 150 Å instead of 200 Å.
15. p. 73: Co/50 Å instead of CoO/50 Å.
16. d_{Au} denotes Au thickness, not exchange bias as written in p. 83: *...and the dependence of exchange bias (d_{Au})...*
17. Too small figure - it is hard to read axis labels in the inset of Fig. 6.4 b in lower panel.
18. Verb used twice, p. 85 : *...H_{EB} drops down decreased to zero...*
19. Numerous mistakes in the References section:
   - there is not enough information in the citation of books in Refs.: 12, 20, 21, 28, 31, 35, 90, 100, 114, 126;
   - lack of page number or volume in Refs.: 40, 50, 57, 67, 68, 71, 72, 116, 118, 120, 122, 124, 125, 128, 129, 130, 132, 133, 134, 137, 142, 147, 149, 150, 152;
   - no author/authors and other information in Ref. 48.

A more general remark concerns the citation of papers that Hashim Nayyef is a co-author of and whose results are presented in the dissertation. In particular, this applies to figures included in the doctoral thesis, which are either identical or slightly modified compared to those in the publications. In my opinion the citation should be included in all relevant figure captions as well as in the body of the dissertation. The citations of the published articles should be given in all places in accordance with the rules applicable to scientific articles.

Finally, it is worth noting that all experiments were performed under ultrahigh vacuum conditions and the prepared samples were fabricated using molecular beam epitaxy method. Beside standard experimental techniques like LEED and MOKE the Author carried out experiments using advanced measurement methods: XMLD, XMCD and corresponding microscopies XMLDPEEM and XMCDPEEM using synchrotron radiation. This proves the appropriately broad scope of knowledge and skills of the Author of the doctoral dissertation.
Hashim Nayyef coauthored five papers related to the topic of his dissertation being the corresponding and second author in one paper (Nuclear Instruments and Methods in Physics Research B) and the first author in one of the papers (Scientific Reports). In addition, Hashim Nayyef coauthored four more articles connected to AFM and FM layers. These achievements demonstrate the ability to conduct independent scientific research by Hashim Nayyef.

Summarizing, the dissertation presents numerous important results broadening our knowledge on AFM/FM multilayers, specifically, on the possibilities of tuning magnetic anisotropy of antiferromagnetic materials. It delivers experimental evidences on a reorientation of magnetic moments in AFM materials by various means together with the possible scenarios responsible for the observed changes in the magnetic properties. In particular, the Author proposes different methods to control direction of the magnetic moments in the antiferromagnetic layers which include thermal modification or interface engineering of magnetic anisotropy. One of the most interesting, also from the application point of view, is the magnetic field free, thermally driven ability to achieve two orthogonal states in the antiferromagnetic layer. This makes it possible to use considered multilayer containing antiferromagnetic material for data recording.

To sum up, the doctoral dissertation submitted for review is a homogeneous description of magnetic properties of antiferromagnetic/ferromagnetic multilayers. It delivers new knowledge on the control of magnetic anisotropy and easy axis in antiferromagnets and as that it is an original solution of a scientific problem. Therefore, the doctoral dissertation of Hashim Nayyef meets the requirements specified in art. 187 of the Act of July 20, 2018, Law on Higher Education and Science (as amended) and may be admitted to further stages of the doctoral process.

Ryszard Zdyb