The Henryk Niewodniczański
INSTITUTE OF NUCLEAR PHYSICS
POLISH ACADEMY OF SCIENCES

Dr hab. inż. Małgorzata Kąc
Institute of Nuclear Physics PAN
ul. Radzikowskiego 152
31-342 Kraków

Kraków 29.02.2024

Review of the doctoral thesis of Hashim Nayyef entitled:

Tailoring the magnetic anisotropy of antiferromagnetic thin films in epitaxial multilayer systems and ferromagnetic / antiferromagnetic nanostructures

Magnetic nanostructures enjoy unflagging interest both from the point of view of basic research and due to their application potential. Among them, antiferromagnetic thin films occupy an important place as a material that can meet the requirements of spintronics. Compared to ferromagnets (FM), antiferromagnetic materials are insensitive to external magnetic fields, do not generate stray fields, and exhibit ultrafast dynamics. Despite their significant advantages, antiferromagnets (AFM) have not been intensively studied for many years because the zero net magnetization made them less useful as active components of spintronic devices. Additionally, difficulties in adjusting the magnetic properties of antiferromagnets also did not contribute to the interest in such materials, whose application was usually limited to passive components acting as pinning layers in spin valves. The breakthrough was the discovery of the possibility of controlling the orientation of the Néel vector, which can be done using magnetic, strain, electrical, or optical methods. Since then, there has been an exponential increase in interest in antiferromagnets, with particular emphasis on detecting and manipulating their magnetic ordering.

This challenge was also undertaken by Mr. Hashim Nayyef in his doctoral thesis, in which he presents the studies of the interaction between ferromagnetic Fe sublayer and NiO or CoO antiferromagnetic overlayers. The Author analyzed three systems grown on tungsten (110) substrate composed of the following films: NiO(111)/Fe(110) bilayers, and CoO(111)/Fe(110) bilayers and trilayers, in which magnetic layers are separated by gold layer i.e. CoO(111)/Au(111)/Fe(110) systems. The Author manipulated the magnetic moments of antiferromagnetic and ferromagnetic layers by changing the sublayer thickness and applying temperature or a small magnetic field, which is the goal of this work.

The experimental part of the work was preceded by two chapters. The first one contains a description of magnetism in the analyzed structures with special attention paid to magnetic anisotropy, its types, and origin. The second one includes a description of the sample preparation and its structural and magnetic characterization.

In the first part of the magnetic section, in addition to basic information, there is a clear description concerning in-plane magnetic anisotropy of the Fe(110) layer evaporated on a W(110) substrate with an indication of the spin reorientation transition (SRT) depending on the Fe layer thickness and annealing temperature. This part corresponds to the system analyzed in the experimental studies. The next subsection presents the magnetic properties of antiferromagnetic thin films and describes the interfacial interaction between ferromagnetic and antiferromagnetic layers when they are adjacent and separated by a non-magnetic layer.

The methodology chapter focuses on the description of the molecular beam epitaxy method used for sample preparation and the techniques applied for structural analysis, i.e. low-energy electron diffraction, and magnetic characterization methods such as the magneto-optic Kerr effect, X-ray magnetic circular dichroism, and X-ray magnetic linear dichroism. The appropriate techniques used by the Author allowed him to observe a magnetic ordering in the ferromagnetic layers, as well as in the antiferromagnetic overlayers.

> Question 1. The Author did not mention the X-ray photoemission electron microscopy technique in this chapter. What is the surface resolution of this technique?

The main part of the work, containing the results is described in the next three chapters.

The first concerns NiO(111)/Fe(110) systems. The Author manipulates the magnetic states of antiferromagnet between two orthogonal directions by changing the thickness of the Fe sublayer, as well as by lowering the temperature or applying a small magnetic field, which modifies the interfacial magnetic anisotropy, thermal hysteresis of spin reorientation transition and interfacial exchange coupling. The antiferromagnetic NiO layer, whose spins easily rotate with the magnetization vector of Fe film, turned out to be very sensitive to the magnetic state of the Fe sublayer. A very interesting observation of the presence of two orthogonal directions, which may appear depending on the pathway taken to reach the given temperature, allows the selected antiferromagnetic orientation to be stabilized by heating or cooling the sample in a specific temperature range. It shows the potential application of such structures in heat-assisted magnetic recording technology.

> Question 2. In his work, the Author analyzed the influence of the ferromagnetic layer on the magnetic state of antiferromagnetic oxide. The simulation predicted the changes in the magnetic ordering of the FM layer from [1-10] to [001] depending on the layer thickness. Did the Author notice the influence of the AFM overlayer on the critical thickness of the spin reorientation transition? In other words, does the spin reorientation transition in the Fe layer not covered by AFM also occur at the same Fe layer thickness?

The next chapter, dedicated to the studies of CoO(111)/Fe(110) systems, focuses on the investigation of the exchange bias and interfacial spin orientation. The Author found that the magnetic state of the Fe layer achieved above the Néel temperature of the CoO layer may determine the magnetic orientation of the antiferromagnetic layer. This magnetic configuration of CoO remains frozen even when the orientation of the ferromagnetic layer changes. On the other hand, the state of antiferromagnet imprinted by ferromagnet affected the strength of magnetic anisotropy of FM at low temperatures. Unlike the previous system, the spins in CoO were not fully rotatable (as in NiO), which can result from significantly smaller magnetic anisotropy of NiO compared to CoO. Another important difference is the strong exchange bias (EB), which indicates the spin orientation of the antiferromagnetic layer. Appropriate cooling of the system allowed the Author to manipulate the axis and direction of interfacial antiferromagnetic spins.

In the third system, in which the CoO(111)/Fe(110) bilayer is separated by the thin gold layer, studies of effective anisotropy of the ferromagnetic layer and strength of the exchange bias are presented. The Author noticed interesting, non-monotonous behavior of EB as a function of the spacer thickness with an enhancement of about 100% (compared to systems without spacer) at the gold thickness of 1 monolayer (ML). Moreover, it was found that the magnetic state of the ferromagnetic layer was determined by the antiferromagnetic layer, which as long as the thickness of the spacer allows for AFM-FM interactions, induces [001] orientation of the Fe easy axis. The modification of the magnetic state of the FM and AFM was achieved by variation of Fe and Au layer thicknesses as well as by temperature changes.

> Question 3. What could be the reason for the observed exchange bias enhancement? The Author mentioned rough interfaces, reduction of magnetic frustration, quantum well states, and Fe oxidation suppression. Some of them are excluded by the Author, which one is the most probable? An increase in the exchange bias is observed until the gold layer reaches the thickness of 1ML. Can this layer be continuous, taking into account the surface roughness of the Fe layer and the type of Au growth on Fe(110)? Do the Author's observations agree with the literature explanations [152] indicating that the enhancement of exchange bias is caused by a dusting inserting layer, which reduces the magnetic frustration at the interface? What can be the influence of Au atoms? Can they block the interdiffusion between neighboring layers - smoothening interfaces, or rather diffuse into the FM or AFM layers increasing the interfacial roughness?

The work ended with the conclusions, which clearly summarized the obtained results and highlighted the different nature of studied systems from rotatable NiO spins, switched by a small external magnetic field or an appropriate temperature and Fe thickness selection to frozen spins in CoO, which using FC procedure were stable below T_N, even when the magnetic state of Fe changes. The latter system was additionally modified by the spacer introducing, which extended the spin manipulating possibility between various orientations of both ferromagnetic and antiferromagnetic layers through the appropriately selected thickness of the ferromagnetic layer and non-magnetic spacer as well as by temperature adjustment.

The doctoral thesis is compact and consists of 103 pages with 152 references and a list of nine articles co-authored by Mr. Hashim Nayyef. The structure of the work is clear and the subsequent chapters are arranged logically. The aim of the work, which was to manipulate the magnetic state of the antiferromagnetic layer, was achieved. The Author, who has undoubted knowledge of this topic, presents information on the issues discussed in the experimental part based on extensive literature studies of articles with high impact factors. Mr. Hashim Nayyef also demonstrated very good knowledge of the techniques used, which were exceptionally well-selected and allowed him to study both ferromagnetic and antiferromagnetic materials. In the experimental part of the work included in the next three chapters, the Author presents the results, showing carefully prepared figures along with their comprehensive description and in-depth discussion. Their high quality is proven by the fact that all the results were published in journals with a high impact factor. The innovative nature of the topic and the application potential of the presented results are also noteworthy.

The thesis contains several editorial shortcomings, which in no way affect my very high assessment of the reviewed work, but as a reviewer, I am obliged to mention them. I would like to ask Mr. Hashim Nayyef to answer the three questions included in the review and briefly comment on the five underlined items from the list below regarding the missing information.

1. Year of publication Ślęzak et al. entitled *Tailorable exchange bias and memory of frozen antiferromagnetic spins in epitaxial CoO(111)/Fe(110) bilayers* is incorrectly reported (2021 instead of 2022).
2. Acronyms (MA, AFM, FM) should be explained in the main text not only in the abstract. A list of used acronyms at the beginning of the work would be helpful.
3. The critical thickness for switching the easy direction of magnetization is about 110 Å in the discussed studies taken from reference No. 45 while the Author, quoting this paper, pointed out the critical thickness below 100 Å (Fig. 2.6, p. 24).
4. The book (ref. No. 48) is listed in the Reference section without the bibliographic data.
5. On pages 34-35 and 35-36, information on the impact of volume defects on interface coupling has been duplicated.
6. Decay length value taken from ref. No. 89 is misquoted as 0.5 Å, while the value of 2 Å is given in the paper (p. 37).
7. The acronym PIRX is not explained.
8. The description of the techniques used lacks information regarding sample preparation for measurements. It would be interesting to know the sensitivity of these methods and the sample sizes required to obtain a measurable signal.
9. A LEED pattern of the W(110) surface could be shown with a description indicating the mutual crystallographic orientations and lattice mismatch enabling epitaxial growth of Fe(110) layers. Additionally, the Author did not comment on the growth of the NiO (111) layer on the Fe(110) surface (Fig. 4.1 e).
10. The scale bar description is missing in Fig. 4.4 c. p. 57.
11. It is not clear if the results presented in Fig. 4.10 of the top panel were obtained for Fe layer thicknesses of 50 Å, 90 Å, and 150 Å or 85 Å, 90 Å, and 95 Å as shown in the publication (p. 66).

12. The Author refers to the easy axis of magnetic anisotropy in the bulk materials, it would be useful if the easy and hard axes of bulk oxides were indicated.
13. The "significant difference" between the spectra measured for the two Fe thicknesses is difficult to notice, especially when the spectra were covered with a field cooling line (Fig. 5.1 d).
14. The references number 41 and 138 have been duplicated.
15. A short comment on Fig. 6.2 describing the epitaxial growth of subsequent layers is missing (p. 80).
16. The Author uses the atomic layer term without defining the thickness of the monolayer of Fe and Au.

As I mentioned above, these editorial shortcomings do not detract from the substantive value of the work. Research performed on two types of systems containing NiO and CoO layers, which showed rotating and frozen spins, respectively, proved that the Author can manipulate not only the direction of spin orientation in the antiferromagnetic layer but also the magnetization vector of the interface layer. This is a current topic, crucial for the further development of antiferromagnet-based devices and providing a new perspective on spintronic technology. The presented results, published in five renowned scientific journals, certainly contribute to a better understanding of the behavior of the antiferromagnetic layer in the proximity of ferromagnet that exhibits a thickness-induced in-plane spin reorientation transition.

In light of the above, I consider this work to be extremely valuable, undoubtedly constituting an innovative contribution to the field of antiferromagnetic spintronic devices and I am applying for the distinction to be awarded to this thesis.

I declare that the doctoral dissertation entitled *Tailoring the magnetic anisotropy of antiferromagnetic thin films in epitaxial multilayer systems and ferromagnetic/antiferromagnetic nanostructures* meets the requirements of The Law on Higher Education and Science, Act of 20 July 2018, Art. 187 as amended and I am asking for consent to the public defense of Mr. Hashim Nayyef, M.Sc., before the Scientific Discipline Council for Physical Sciences of the AGH University of Science and Technology.

Małgorzata Kąc