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A review of the doctoral dissertation of Mr. Sukanta Kumar Jena, M.Sc., entitled "Magnetic properties of asymmetric layered heavy metal/ferromagnet system"

The doctoral dissertation of Sukanta Kumar Jena, M.Sc., was made at the Institute of Physics of the Polish Academy of Sciences in Warsaw under the direction of Prof. dr hab. Andrzej Wawro *[professor, Ph.D., postdoctoral degree holder]*, whose assistant supervisor was dr Ewelina Milińska *[Ph.D.]*. In the further course of my review, let me refer to Mr. Sukanta Kumar Jena as the Candidate for purely stylistic reasons related to the difficulty in inflecting his name in Polish.

Following the title of the dissertation, it concerns the study of the magnetic properties of thin-film structures based on asymmetric sequences of thin ferromagnetic (Co) layers in contact with layers of heavy metals – tungsten (W) and platinum (Pt). Thus, W/Co/Pt sequences with a thickness of several to several tens of angstroms (Å) and with n repetitions of these sequences in the range from 1 to 20 are the subject of the Candidate's research. Magnetic layer structures have been the subject of intensive research for about 30 years, and the most spectacular result of this research was the discovery of the giant magnetoresistance (GMR) resulting from the antiparallel magnetic order in ferromagnetic sublayers separated by "spacers" – layers of non-ferromagnetic metals. Another significant research achievement in this field was the observation of perpendicular anisotropy in Au/Co/Au multilayer structures and the explanation of this effect (Bruno model and X-ray magnetic circular dichroism *[XMCD]* experiments) as a result of the spin-orbit interaction in structures containing symmetry-broken interfaces of the crystal lattice. In this context, the natural development of research on multilayer structures seems to be to check how "asymmetric" structures containing various heavy metals (e.g. W/Co/Pt) modify their effective (i.e. global) magnetic properties related to the perpendicular magnetic anisotropy

(PMA). An important additional effect of such an asymmetric configuration is the Dzialoshynskii-Moriya interaction (DMI), which modifies the spin structure of these structures, leading to the formation of skyrmions.

Sukanta Kumar Jena, M.Sc., is the co-author of two papers published in peer-reviewed journals: *Nanoscale* (first co-author) and *Journal of Magnetism and Magnetic Materials* – second co-author. In the first publication, the Candidate was crucially involved in: sample preparation, MFM measurements, and the analysis of the results, the analysis of the SQID results, the determination of D_{eff} from K_{ef} effective anisotropy measurement data, and in the preparation of the draft version of the manuscript. In the second publication, the Candidate described his contribution as research – investigation. I understand that the Candidate has performed the basic magnetic tests necessary to determine the magnetic order of the Pt/W/Co/Pt structures. Other achievements of the Candidate include: obtaining a doctoral scholarship in the Beethoven project, obtaining funding from the IEEE Magnetic Society, participating in five poster presentations and five oral presentations at a number of national and international conferences, and giving two seminars at the Institute of Physics of the Polish Academy of Sciences and the University of Bialystok.

The doctoral dissertation, written in English, includes 113 pages and consists of six chapters. In addition to the usual bibliographic information on the Candidate, acknowledgments, and introduction, the dissertation consists of two parts. The first describes the theoretical foundations of magnetism and the experimental techniques used in the research (Chapters 2 and 3). The second – covering chapters 4, 5, and 6 – directly concerns the results of research on asymmetric W/Co/Pt thin-film structures and ends with a concise summary. The structure of this doctoral dissertation is therefore typical for works presenting the results of experimental research in physics.

The introduction to the dissertation contains basic information on the complexity of interactions in metallic multilayer structures and the possibility of their modification by changing the thickness of individual sublayers, changing their sequence, and the number of repetitions of basic structural elements, which are ultra-thin epitaxial W/Co/Pt triplelayers. Thus, these are mainly geometric modifications resulting from the technology used – molecular beam epitaxy (MBE). As emphasized by the Candidate, this technology enabled obtaining structures of a quality unattainable to obtain ultra-thin layers using other methods of deposition. The interactions of interest in this dissertation are primarily the DMI interaction accompanying the perpendicular anisotropy, interlayer coupling, and finally magnetostatic interactions in the case of structures with a significant number of repetitions of the W/Co/Pt trilayers. These interactions (with a significant contribution of DMI) lead to the formation of a specific domain structure evolving into a network of skyrmions.

A characteristic feature of the briefly discussed introduction is the statement about the Candidate's role in the work related to the implementation of his doctoral dissertation. Instead of the usual statements of the co-authors of the publication, the Candidate personally presented his participation in individual studies. I believe that such an approach is equal and possible comments may appear during the public defense of the doctoral dissertation.

Chapter 2 contains condensed, textbook information on the basic phenomena related to ferromagnetism with particular emphasis on the anisotropy of thin films, DMI, and domain structure. In most doctoral dissertations in the field of magnetism, the authors usually include a chapter devoted to theoretical foundations. I think this chapter might as well be skipped, especially since basic information can be found on the Internet these days. The candidate followed the usual path, but he did not avoid a mistake. On page 30, in the last sentence, he stated that for thin layers with perpendicular anisotropy, the demagnetization coefficients Nx=Ny=0 and Nz=1. Such values of demagnetization coefficients are valid for all thin magnetic layers and the perpendicular anisotropy has no relation to it.

Chapter 3 of the dissertation describes the experimental techniques used by the Candidate in the study of epitaxial W/Co/Pt structures. The most detailed presentation is given to the process of depositing samples using the molecular beam epitaxy (MBE) technique for epitaxial layered structures, in which the high quality of the crystal structure and the low roughness of the interfaces have a significant impact on the repeatability of the results and the values of some parameters determined in the experiment. There is a thought-provoking lack of description of the wideband ferromagnetic resonance (VNA-FMR) technique, which the Candidate mentions a couple of times in the fragment regarding grants, scientific visits, and conference presentations. Since the further fragments of the text of the dissertation also do not contain any information about the results obtained from VNA-FMR measurements, I conclude that the results of these studies were either of little importance or were not developed more thoroughly. Therefore, I would ask the Candidate to comment on this subject during the public defense.

The fourth chapter describes the crystallographic structure of the produced multilayer systems, the magnetic properties of the Co layer in selected types of heterostructures, the effect of contact with tungsten (W) or Pt layers on Co magnetism and the influence of W layer thickness on Co magnetism. Since the 1990s, it has been known that thin layers of Co with a thickness of less than ~18 Å in contact with gold (Au) exhibit perpendicular magnetic anisotropy (PMA). For d_{Co}>18 Å the anisotropy is already of the in-plane type, and such a transition between these types of magnetization ordering is referred to by the term spin reorientation transition (SRT). SRT depends on the type of heavy metal. For example, for the Pt/Co/Pt structure, SRT occurs at d_{Co}=22 Å, and for the Mo/Co/M structure, SRT is absent and the anisotropy is of the easy plane type.

Since Mo and W have a bcc structure and similar lattice constants, the Candidate decided to investigate how the perpendicular anisotropy is realized in W/Co/Pt structures, in which contact with Pt favors the occurrence of PMA, while contact with W should (as with Mo) lead to the absence of PMA. The Candidate studied a whole range of W/Co/Pt structures in various configurations and came to interesting conclusions that W/Co/Pt structures are significantly different from Mo/Co/Pt structures. In particular, he concluded that the type of buffer layer (Pt or W) determines the presence or absence of PMA in a certain range of Co thickness (Fig. 4.2). To further investigate the effect of the buffer layer, the structural changes that the buffer layer undergoes as it grows over a 400 Å thin Pt layer were observed. RHEED, TEM, and AFM techniques were used for this purpose. The main result of these studies was to show that the W layer (for d_W<5 Å) pseudomorphically grows in the fcc structure and then epitaxially in the bcc structure, forming crystallites oriented in three directions relative to the main crystallographic directions of the (111) Pt plane. The effect of the thickness of W on the perpendicular anisotropy of Co in Pt/W (d_W)/Co(d_{Co})/Pt(40 Å). The results of these tests are summarized in Fig. 4.6, which clearly shows that the d_{Co} range of PMA occurrence is independent of the thickness of W and is evenly shifted towards higher d_{Co} values with the increase of the thickness of W. In particular (Fig. 4.6 b) visualizes a spectacular map of various magnetic states (configurations) occurring in the entire tested range of d_{Co} versus d_W. I consider this result to be a particularly important element of the doctoral dissertation, especially since it must have required many burdensome measurements.

Further in Chapter 4, the Candidate discussed the results of the anisotropy field measurements and discussed the effect of the asymmetric W/Co and Co/Pt interfaces on the thickness of d_0, the so-called dead layer. An interesting and unexpected result for me is the occurrence of a negative d_0 value for small thicknesses of dW<20 Å in the Pt/W(d_W)/Co(18 Å)/Pt system. It turns out that this is due to two opposite effects: the polarization of Pt atoms at the Co/Pt interface causing an increase in the effective magnetic moment, and the quenching of the magnetic moment at the W/Co interface. The result of these opposing effects is the non-monotonic nature of the d_0 vs. d_W relationship (Fig. 4.9 b). This relationship would require a more thorough discussion. In particular, explaining why these effects compensate (giving d_0=O) at d_W=40 Å, may contribute to the modeling of the structure of the W/Co and Co/Pt interfaces.

The discussion of the domain structure of the W/Co/Pt structures is descriptive and I do not feel competent to evaluate this discussion. The most important result of this part of the dissertation is the observation of a fine structure – magnetic skyrmion bubbles – the realization of which is attributed to the presence of a significant iDMI impact on the interfaces. The Candidate determined the value of the interaction constant D-M: D=-1.12 mJ/m^2 using Brillouin spectroscopy.

In conclusion, Chapter 4 provides several interesting results on the anisotropy and magnetic interactions of asymmetric W/Co/Pt structures with particular emphasis on the perpendicular anisotropy, the unusual nature of the so-called dead layer, domain structure evolution, and D-M interface interaction. Unfortunately, the "narrative" style of this chapter made it difficult for me to obtain a comprehensive picture of the phenomena responsible for the observed effects. Also, some illegible drawings cause difficulties for the reader.

The fifth chapter deals with the study of the remagnetization processes of single W/Co/Pt layers and more complex structures consisting of 6, 7, and 10 repetitions of W/Co/Pt blocks. For single W/Co/Pt structures, based on local measurements of the Kerr loops in specially prepared samples with orthogonal wedges of W (0-20 Å) and Pt (0-19 Å) plotted in the form of sequential steps, it was possible to visualize changes in the shape of the hysteresis loop in the form of a two-dimensional map. Such a skillful combination of sophisticated technology of epitaxial structures and local PMOKE measurements deserves attention. The main result of these studies was the observation of the interlayer antiferromagnetic exchange coupling for a certain range of thicknesses of the W+Pt sublayers and the determination of the value of this coupling (Fig. 5.4). I highly rate the above results despite the lack of detailed discussion of the nature of the dependence of this coupling on the thickness of d_{W+PT}.

Remagnetization processes in multiple structures are more complicated due to the interaction of RKKY-type interactions (leading to inter-layer exchange coupling), dipole interaction, and the presence of iDMI. The discussion of remagnetization processes in these structures is interesting. Due to interlayer antiferromagnetic coupling, these systems behave like compensated (for even repetitions) or uncompensated (for odd repetitions) artificial antiferromagnets that sequentially remagnetize. The domain structure in the remanence state confirms this global antiferromagnetic nature of the magnetic ordering. Finally, for structures with the thickest W+Pt = 20 Å spacers, for which the interlayer exchange coupling is strongly reduced, the role of dipole interactions increases, as manifested by the specific shape of the hysteresis loop (Fig. 5.7), and the labyrinthine domain structure in the remanence state (Fig. 5.8).

The sixth chapter presents the results of research on the remagnetization processes, and the domain structure of the $[W(10\ Å)/Co(6\ Å)/Pt(10\ Å)]_N$ layers with the number of repetitions N=10 and 20 is characterized in detail. In such multilayer systems, there is both a strong dipole interaction and a significant iDMI, while the exchangeable interlayer coupling is reduced. Based on the observed labyrinthine or striped domain structure and the so-called K_{eff} procedure adapted from the literature (Legrand et al.), the value of the D-M interaction constant was estimated: D=2.65 mJ/m^2. This high value was confirmed by the results of micromagnetic simulations and confronted with numerical calculations from first principles (DFT). In the discussion, the high value of the D parameter was commented on as the result of the perfect structure of W/Co/Pt interfaces with low roughness, which is unattainable for similar layers applied by sputtering.

The Candidate's doctoral dissertation submitted for review, despite critical remarks concerning mainly the manner of editing the text and minor imperfections in the presentation of individual issues, represents a valuable contribution to a fragment of knowledge about complex magnetic thin-film structures that is interesting from the point of view of the mechanisms of magnetic interactions. The author has analyzed in detail the conditions leading to the formation of the entire spectrum of magnetization configurations resulting from the significant contribution of Dzialoshynskii-Moriya type interactions and the realization of the specific domain structure, including the skyrmion lattice. In conclusion, I would like to state that the reviewed dissertation meets the requirements of the Law on Academic Degrees and Titles, and I request that Sukanta Kumar Jena, M.Sc., be admitted to the further stages of the doctoral dissertation.

Janusz Dubowik Poznań, 8.05.2023

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