Deliverable D4.2

Report on metallic multilayer systems

Project number	101070290
Project name	Nonlinear Magnons for Reservoir Computing in Reciprocal Space
Project acronym	NIMFEIA
Work package	WP4 – Materials and device fabrication
Type	Report
Dissemination level	Public
Lead beneficiary	Johannes Gutenberg University Mainz
Due date of delivery	Month 30

Authors: Gerhard Jakob (JGU), Mathias Kläui (JGU)

Disclaimer:

The NIMFEIA project has received funding by the European Union's Research and Innovation Programme Horizon Europe under grant agreement No 101070290. However, views and opinions expressed in this document are those of the authors only and do not necessarily reflect those of the European Union. The European Union cannot be held responsible for them.

1. Development of perpendicular anisotropy materials for high frequency dynamics based on metallic multilayers

1.1. Starting point and literature survey

The standard material for thin film applications exploiting perpendicular magnetic anisotropy (PMA) are Co/Pt multilayers. This combination has been investigated already 35 years ago for possible application in magneto-optical recording [1]. Nevertheless, with refined deposition technologies a continuous optimization process is ongoing, e.g., by optimization of the top layer covering the Pt/Co stack, as both interfaces contribute to the total interface anisotropy. In 2017 Dieny and Chshiev published a review article on the influence of top oxide layers on the PMA with over 300 references [2]. This article is not the endpoint of the story, as demonstrated e.g. by the MgO deposition optimization reported by Gweon et al., leading to saturation fields more than 4T for the hard axis direction [3].

However, Co/Pt multilayers are not best suited for the purposes of NIMFEIA, as Co films provide rather high pinning and high magnetic Gilbert damping. The Gilbert damping of CoFe alloys is lower than that of the respective pure materials as demonstrated by Schön et al. [4] and us [5]. However, the PMA of such alloys and CoFeB systems depends strongly on the respective interfaces. Furthermore, deposition parameters cannot be easily transferred from one deposition system to another. In our Singulus Rotaris deposition tool, we achieved PMA with Pt/$Co_{60}Fe_{20}B_{20}$ whereas the iron rich composition showed PMA only in combination with a tungsten underlayer W/$Co_{20}Fe_{60}B_{20}$ [6]. With respect to damping, permalloy, a NiFe alloy, seems to be the best metallic material. Simultaneously, permalloy is a magnetically extremely soft material, and the influence of remaining pinning sites is seen in the domain formation in Kerr microscopy [7]. Therefore, we investigate Ni-based PMA materials e.g. Co/Ni multilayers [8].

1.2. Strategy and Methods

As written in the proposal, the strategy is to use multilayer systems that exhibit PMA and host spin structures, such as skyrmions, in which higher frequency magnons occur. We will vary heavy metal layers, oxide layers, and magnetic layers as well as deposition conditions and post deposition annealing. The deposition method will be always using the technologically relevant physical vapor deposition tool at JGU that enables deposition on 200 mm wafers and parameters will be transferable to the 300 mm tools at partner GF. For the layer design, we want to minimize spin pumping effects at the interfaces to the heavy metals as those lead to enhanced damping [9]. It is important to note that not only the nominal layer stack is relevant but also the deposition parameters and the post annealing procedures, as they influence the microstructure of the samples. Higher deposition rates, for example, lead to a higher supersaturation of the vapor phase. At initial growth phase, this will lead to a higher nucleation density of film grains and, accordingly, to grains with smaller lateral extension. On the other hand, stable plasma conditions are needed, which limits the range of power settings. With the gas flow in the chamber, one influences the number of collisions of the sputtered atoms in the gas phase and one expects lower energy particles arriving at the substrate for higher gas flows. Another important aspect in our strategy is the use of dusting layers. Reproducible deposition of material layers with a nominal thickness of less than a monoatomic layer is now a state-of-the-art method to fine tune layer properties.

As both low pinning magnetic layers and low damping systems require very homogeneous magnetic systems, we can utilize our experience for optimum skyrmion systems, for both purposes. In order to tailor the size of magnetic structures by their effective anisotropy, we have engineered a magnetic multilayer from a Ta/CoFeB-based stack that hosts Néel skyrmions. Starting from this platform, we have tuned the relative strength of the dipolar interactions by increasing the number of repetitions, n, from n = 1 to 30. The composition of the stack is Ta(4)/[$Co_{20}Fe_{60}B_{20}$(0.86)/Ta(0.06)/MgO(2)/Ta(2)]×n (layer thicknesses in nm with 0.01 nm precision), with n being the number of repetitions. Tey were deposited using a Singulus Rotaris magnetron sputtering tool with a base pressure of 3 × 10^{-8} mbar onto Si/SiO_2 substrates. We propose an analytical model for the skyrmion size and period as a function of the number of repetitions, which we expand to the limit of $n \to \infty$ under the assumption of a vertically homogeneous magnetization. Our results demonstrate the opposite scaling with dipolar coupling with respect to isolated skyrmions, enabling a way to increase skyrmion density by increasing the dipolar coupling, which also favors the robustness of spin textures against external magnetic fields. Note that at the lower limit ($n \to 0$), our model reproduces the same scaling as the one already described for stripe domains [10-13], with a different prefactor, while the upper limit ($n \to \infty$) reproduces the same scaling of the Kittel model for one-dimensional domain walls [13]. We could also describe the nucleation procedure of the skyrmion lattice starting from a stripe situation, elucidating the key role of the starting stripe periodicity (i.e., energy minima) in the final skyrmion lattice state (overfilled lattice vs. not overfilled). Our calculations yield excellent agreement with the experiments, as well as with the results from micromagnetic simulation, enabling us to fully capture the physics underlying these dipolar-stabilized skyrmion lattices. Details of these investigations are published in [A].

While the above multilayers show high tunability of their magnetic properties and demonstrates the versatility of skyrmion systems, the high heavy metal content will be unfavorable for implementations in magnetic reservoirs. For this purpose, we focused on CoNi multilayers [B]. The final stacks are as Substrate/Ta(3)/Ru(2)/Pt(1.5)/[Co(0.2)/Ni(0.6)]3/Co(0.2)/MgO(2)/CoFeB(0.3)/Ta(1)/Ru(5). The CoNi multilayer provides the strong out-of-plane magnetic anisotropy due to the internal interfaces. We used these systems to determine the strength of orbital torque contributions in addition to spin torque contributions to the switching of the PMA layer. We patterned samples to Hall bar structures as shown in Fig. 1 on the left side. Using measurements of second harmonic contributions to the Hall voltage, while varying the magnetic field direction, we determined the strengths of the field-like and damping-like torques on the PMA multilayers. For applications, the decisive criterion is the switching current, and we found this to be lowered due to orbital torque contributions from the Ru (see right side of Fig. 1). As the magnetization of such multilayers reacts strongly to the currents and not only to external fields, it opens the potential to feed the radar signals to the spin torque layer directly instead of the antenna structures sketched in the original NIMFEIA proposal. Those interact only via the Oersted fields but the direct interaction at the interface promises better scaling to smaller system sizes. However, current densities might be too high using this idea.

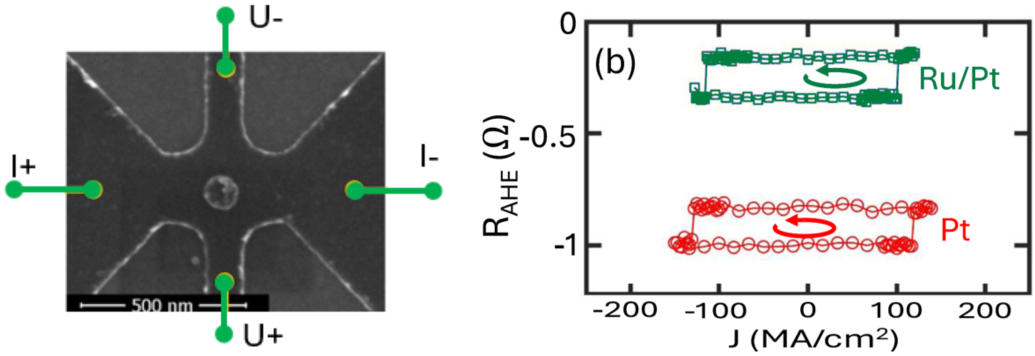

Figure 1: Magnetization switching of PMA multilayer by orbital torques. (a) Scanning electron microscope image with circuit diagram of the switching experiment. (b) Anomalous Hall resistance (R_{AHE}) as a function of current density at a 10 ns pulse duration and $\mu_0 H_x$= 50mT. The y-axis has been rescaled for better visualization, and rounded arrows indicate the direction of switching polarity (Figure modified from [B]).

If one is working with PMA magnetic multilayers, one wants to have a stable defined direction of the magnetic field. For this purpose, exchange bias layers are applied and we found that we can stabilize PMA with strong exchange bias in PtMn/Co by magneto-ionics [C]. We expect that we can transfer this idea also to the Co/Ni-based multilayers where the initial state in previous experiments was always set by a high external magnetic field.

1.3. Summary and conclusions

In conclusion, we investigated CoFe-based multilayers as well as CoNi-based multilayers within task 4.2. More detailed results than mentioned in this report can be found in the scientific papers that are published with NIMFEIA acknowledgements in [A], [B] and [C]. These papers discuss also topics that are outside the scope of NIMFEIA.

2. Publications

[A] **The role of magnetic dipolar interactions in skyrmion lattices,**
Elizabeth M. Jefremovas, Kilian Leutner, Miriam G. Fischer, Jorge Marqués-Marchán, Thomas B. Winkler, Agustina Asenjo, Robert Frömter, Jairo Sinova, Mathias Kläui,
arXiv2407.00539v1 (2024), https://doi.org/10.48550/arXiv.2407.00539

[B] **Harnessing Orbital Hall Effect in Spin-Orbit Torque MRAM Devices**
Rahul Gupta, Chloe Bouard, Fabian Kammerbauer, Omar Ledesma-Martin, Arnab Bose, Iryna Kononenko, Sylvain Martin, Perrine Use, Gerhard Jakob, Marc Drouard, Mathias Kläui,
Nature Commun. 16, 130 (2025), doi: 10.1038/s41467-024-55437-x

[C] **Stabilizing perpendicular magnetic anisotropy with strong exchange bias in PtMn/Co by magneto-ionics**
Beatrice Bednarz, Maria-Andromachi Syskaki, Rohit Pachat, Leon Prädel, Martin Wortmann, Timo Kuschel, Shimpei Ono, Mathias Kläui, Liza Herrera Diez, and Gerhard Jakob,
Appl. Phys. Lett. **124**, 232403 (2024), doi: 10.1063/5.0213731

3. References

[1] W. B. Zeper, F. J. A. M. Greidanus, P. F. Carcia, and C. R. Fincher, *Perpendicular magnetic anisotropy and magneto-optical Kerr effect of vapor deposited Co/Pt -layered structures*, J. Appl. Phys. **65**, 4971 (1989).

[2] B. Dieny and M. Chshiev, *Perpendicular magnetic anisotropy at transition metal/oxide interfaces and applications*, Rev. Mod. Phys. **89**, 025008 (2017).

[3] H. K. Gweon, S. J. Yun, and S. H. Lim, *A very large perpendicular magnetic anisotropy in Pt/Co/MgO trilayers fabricated by controlling the MgO sputtering power and its thickness*, Sci. Rep. **8**, 1266 (2022).

[4] M. A. W. Schön, D. Thonig, M. L. Schneider, T. J. Silva, H. T. Nembach, O. Eriksson, O. Karis, and J. M. Shaw, *Ultra-low magnetic damping of a metallic ferromagnet*, *Nat. Phys.* **12** 839842 (2016).

[5] R. Weber, D.-S. Han, I. Boventer, S. Jaiswal, R. Lebrun, G. Jakob, and M. Kläui, *Gilbert damping of CoFe-alloys*, J. Phys. D: Appl. Phys. **52** 325001 (2019).

[6] S. Jaiswal, K. Litzius, I. Lemesh, F. Büttner, S. Finizio, J. Raabe, M. Weigand, K. Lee, J. Langer, B. Ocker, G. Jakob, G. S. D. Beach, and M Kläui*, Investigation of the Dzyaloshinskii-Moriya interaction and room temperature skyrmions in W/CoFeB/MgO thin films and microwires*, Appl. Phys. Lett. **111**, 022409 (2017).

[7] A. Hubert and R. Schäfer, *Magnetic Domains, The Analysis of Magnetic Microstructures*, Springer Berlin, (1998)

[8] K. Ito, N. Kikuchi, T. Seki, and K. Takanashi, *Perpendicularly magnetized epitaxial Co/Ni multilayers grown on Ru (0001) layers by alternate monoatomic layer deposition*, J. Magn. Magn. Mater. **563**, 169908 (2022).

[9] T. G. A. Verhagen, H. N. Tinkey, H. C. Overweg, M. van Son, M. Huber, J. M. van Ruitenbeek, and J. Aarts, *Temperature dependence of spin pumping and Gilbert damping in thin Co/Pt bilayers*, J Phys Condens Matter **28**, 056004 (2016)

[10] I. Lemesh, F. Büttner, and G. S. Beach, *Accurate model of the stripe domain phase of perpendicularly magnetized multilayers*, Physical Review B **95**, 174423 (2017).

[11] J. Cape and G. Lehman, *Magnetic Domain Structures in Thin Uniaxial Plates with Perpendicular Easy Axis*, J. Appl. Phys. **42**, 5732 (1971).

[12] B. Kaplan and G. Gehring, *The domain structure in ultrathin magnetic films*, J. Mag. Mag. Mater. **128**, 111 (1993).

[13] C. Kittel, *Theory of the Structure of Ferromagnetic Domains in Films and Small Particles*, Phys. Rev. **70**, 965 (1946).