Deliverable D7.2

Report on higher anisotropy and coupled materials leading to higher frequencies

Project number	101070290
Project name	Nonlinear Magnons for Reservoir Computing in Reciprocal Space
Project acronym	NIMFEIA
Work package	WP7 – Towards AFM and THz
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. Higher anisotropy and coupled materials leading to higher frequencies

1.1. Starting point and literature survey

The excitation frequencies for thin ferromagnetic films can be estimated from Kittel's formula for the ferromagnetic resonance and scale with the magnitude of the magnetization and the strength of the anisotropy [1]. As we do not have any material exceeding the saturation magnetization of 3d magnetic metal layers significantly, only the anisotropy can be tuned in a limited range. These efforts within NIMFEIA are reported within Deliverable 4.2. A completely different dynamics evolves for antiferromagnetic materials. Here the exchange interaction sets a field scale of hundreds of Teslas and leads to dynamics that are two to three orders of magnitude faster, reaching the THz regime [2]. Information on antiferromagnetic magnons of such high energy has been gained for long time mainly by light and neutron scattering [3] as direct high frequency electronics was not available. However, the advent of improved THz radiation sources and detectors and the refining of electronic equipment is closing this THz gap. A narrowband THz radiation emission has been observed recently from antiferromagnetic magnons with participation from JGU [4]. For generic antiferromagnets, many possess magnon modes in the high THz regime, so we will focus on those antiferromagnets that have resonances in the sub-THz regime but still much higher than any ferrimagnet. In exploiting resonance modes in synthetic antiferromagnets, one might get a useful compromise. The RKKY interaction couples two ferromagnetic layers antiferromagnetically and is less strong than the direct exchange interaction. Thus, higher frequency dynamics of the coupled system is expected than for a pure ferromagnet. At the same time, the individual layers can be manipulated by external disturbances allowing easier probing of the system than for antiferromagnets. A recent review on magnon modes in ferromagnetic and antiferromagnetic systems is provided by Chen et al. [5], including 200 citations of relevant work.

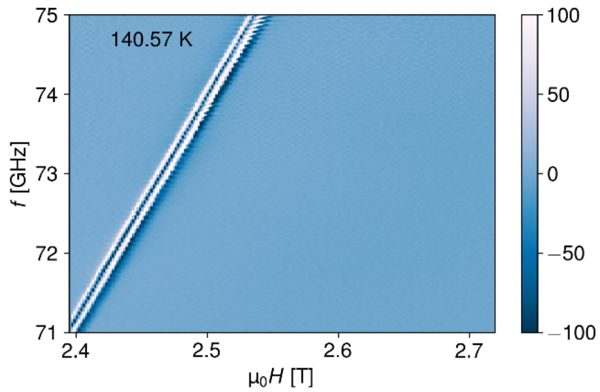
1.2. Strategy and Methods

With respect to higher frequency materials, we mainly looked for oxides where long-distance spin transport has been shown or is expected. This long-distance spin transport must be associated with low damping, a prerequisite to achieve the nonlinear response of the magnetic reservoir based on these materials. Simultaneously, we extended the frequency range of our ferromagnetic resonance setup to 70-90 GHz. Only using such a state-of-the-art electrical detection, we will be able to directly probe excitation spectra of antiferromagnetic magnons by electric means directly.

Concerning the upgrade of the experimental setup, we acquired subharmonic mixers, used as sending and receiving unit. The sending module is provided with two microwave signals f_1 and f_2. The resulting frequency $f = f_1 \pm k \times f_2$ is a mixed signal of these two frequencies, while one of these frequencies (f_2) is lifted to a higher harmonic k, in our case $k = 8$. The modules have a WR12 rectangular waveguide output, allowing us to reach frequencies between 70 to 90 GHz.

One crucial step towards the development of the setup was to fabricate a cryostat insert to mount a sample inside a cryostat with a superconducting coil. For this, we had the need of custom, 2 m long WR12 waveguides, which are attached to custom holders and a waveguide to coplanar waveguide transition, to be able to measure broadband (70-90 GHz). We use two microwave synthesizers for the LO of the mixers and a VNA, connected to the IF port.

The first test measurement was conducted on a LPE grown YIG (3 um) grown on GGG (111), which shows the Kittel Mode at high fields and frequencies. Two modes are observed for this particular sample, as it appears not fully epitaxially grown, possibly in two phases.



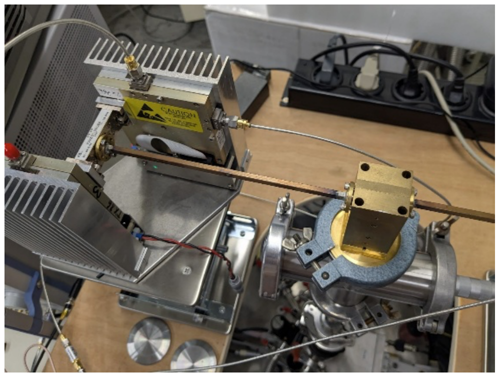


Figure 1: (left) Broadband VNA-FMR measurement of LPE grown YIG, at 140.5 K using the 70-80 GHz module. The raw data is processed by derivative divide. (right) 70-80 GHz and 80-90 GHz sending units, connected to the head of the sample insert on top of the cryostat. The connection is done with WR12 rectangular waveguides.

Concerning the materials investigated, we worked on the antiferromagnets Fe_2O_3, NiO, CoO, $TmFeO_3$, and the ferrimagnetic garnet $Y_3Fe_5O_{12}$ (YIG) that we coupled to the garnet $Gd_3Fe_5O_{12}$ (GdIG). In the latter compound the Gd magnetic moments behave superparamagnetic and align antiparallel to the exchange field created by the iron atoms in tetrahedral and octahedral environments. The Fe ions in the different environments show a ferrimagnetic spin alignment themselves [6]. The temperature-dependent net magnetization that vanishes at the magnetic moment compensation temperature, which is in the bulk of $T_{comp} \approx 295$ K [7]. The investigation of the dynamics of ferrimagnets close to their compensation temperature has been challenging [8,9]. However, coupling to a second layer can potentially be used to facilitate such studies. We experimentally investigated YIG/GdIG/Pt thin film heterostructures (YIG 36 nm/GdIG 30 nm/Pt 4 nm). We observed a strong impact of the magnetic configuration of the individual layers of our heterostructure on the spin dynamics, spin pumping and the spin Seebeck effect signals. We could show that the generated spin current originates from the GdIG layer, which gives us the unique opportunity to investigate the GdIG spin dynamics individually, aided by the coupling to the YIG layer, close to the compensation temperature. This temperature range is usually difficult to study because of the diverging line width of single GdIG layers at the compensation temperature [8]. We have driven the ferromagnetic resonance (FMR) modes of our heterostructure and have measured the spin current which is pumped across the GdIG/Pt interface when a resonance condition is satisfied [1]. We detected this spin current from the spin pumping (SP) by means of the inverse spin-Hall effect (iSHE) in the Pt top layer. We obtained unique information about the switching behavior of our GdIG layer by observing the spin Seebeck effect (SSE) [10]. We compared these results with SSE measurements, in which the gradient is generated by microwave heating during the SP measurements [11]. The microwave-induced SSE is a key tool to determine the switching of the top GdIG layer during the SP measurement itself and is less susceptible to temperature mismatch compared to remounting the sample in another setup. In the FMR measurement of the heterostructure we can detect a high frequency mode in addition to the low frequency YIG mode. Both modes do not behave as one would expect from the Kittel equation for in-plane applied external magnetic fields for single layers, which we attribute to their coupling. The coupled dynamics of the system are especially of interest for further studies and applications exploiting coherent spin transport. Details of these investigations are published in [A].

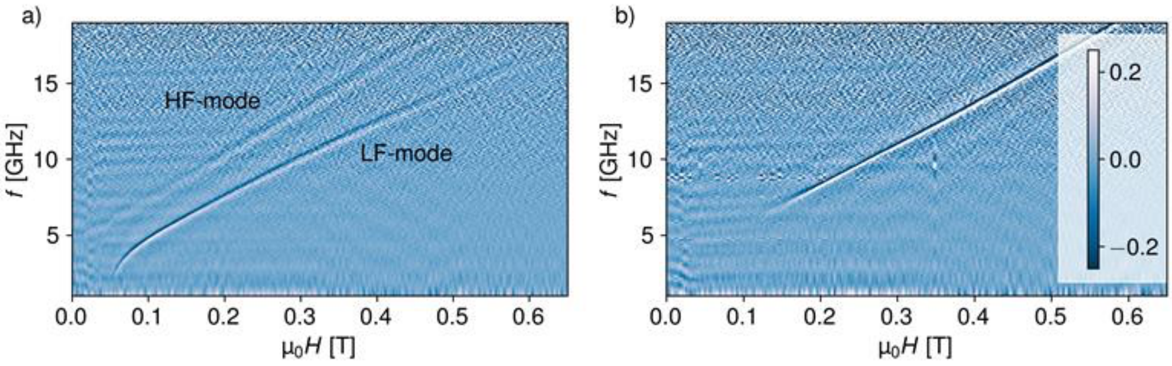


Figure 2: Broadband VNA-FMR measurement of the YIG 36 nm/ GdIG 30 nm/ Pt 4nm trilayer at a) 50 K and b) 200K. The raw data is processed by derivative divide and an FFT-filter. (Figure modified from [A]).

Another system where we want to exploit the coupling between a fast antiferromagnet and a slower ferrimagnetic system is the combination of the iron oxides hematite Fe_2O_3 and magnetite Fe_3O_4. Deposition of these material systems is done in cooperation with university of Bielefeld using molecular beam epitaxy in a controlled oxygen atmosphere. The growth of the material can be monitored in-situ by electron diffraction and ex-situ by x-ray diffraction and x-ray photoelectron spectroscopy. As the materials differ only in the oxygen content and the unwanted phase maghemite is even more stable at high temperatures, this is an ambitious task. Nevertheless, the bilayer system has recently been successfully grown by us and we plan to investigate its spin dynamics within NIMFEIA.

In NiO films, we had earlier found that antiferromagnetic domains can be switched due to thermomagnetoelastic coupling [12]. Dynamic strain could thus be used to control antiferromagnetic states and potentially the AFM dynamics on ultrafast timescales. Using this approach, one can envision to generate THz magnons using ultrafast strain gradients to achieve a time-dependent modulation of the exchange interaction. In [4], in cooperation with other groups, we reported the combined generation of narrowband coherent THz emission centered at 1 THz and incoherent broadband THz magnons in NiO/Pt bilayers. This frequency, however, is too high to be detected directly with electric means in our lab and, thus, these excitations cannot be directly exploited within NIMFEIA.

With our frequency extended FMR setup, however, we could successfully probe two different antiferromagnetic oxides hematite and the orthoferrite $LuFeO_3$. Hematite has a hexagonal crystal structure and orders antiferromagnetically with the easy axis along the c-axis at low temperatures as shown in Fig. 3. Above the Morin transition temperature, the spins orient in the ab-plane and show a slight canting. For bulk crystals, the Morin transition temperature is 260K and we demonstrated long-distance spin transport in this antiferromagnet. Due to its low damping we planned to investigate its magnon modes directly using electrically excited magnetic resonance. While these measurements are ongoing, Fig. 3 shows some preliminary data of the transmission spectrum. At fields near 3 T, we can clearly identify the magnon dispersion curve of the antiferromagnet, which is decreasing with increasing magnetic field for the lower mode [2] (bottom left in Fig. 3).

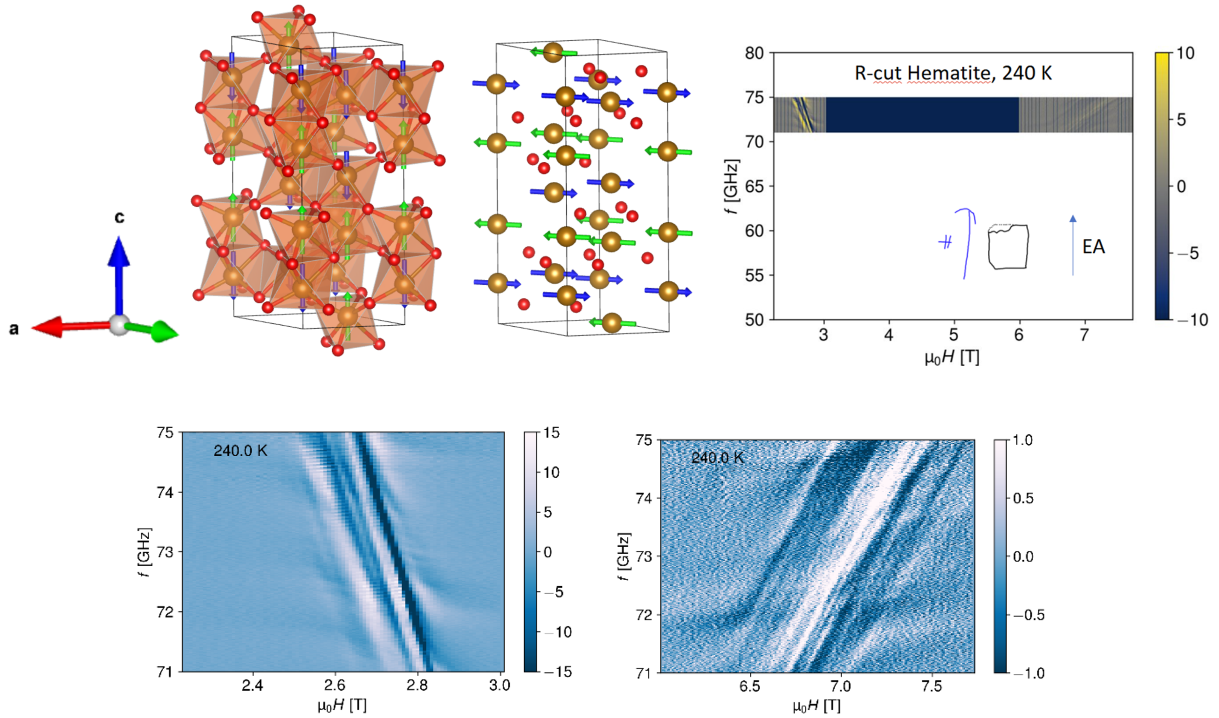


Figure 3: Crystal and spin structure of hematite below (top left) and above (top center) the Morin transition temperature. In the top left panel, additionally the octahedral environment of the Fe atoms is highlighted. Image generated by software VESTA [13]. The top right panel shows preliminary measurements of the resonance absorption. The antiferromagnetic resonance mode at low fields is zoomed in at bottom left, while the quasi-ferromagnetic mode above the spin-flip transition is shown enlarged at bottom right.

For the orthoferrite $TmFeO_3$ the underlying crystal structure is orthorhombic. The structure is derived from the cubic perovskite structure and ordered tilts and rotations of the oxygen octahedra lead to a larger unit cell of lower symmetry. We were motivated to check spin transport in this material due to our finding of long-distance spin transport in the orthoferrite $YFeO_3$. [16]. The rare earth (RE) substituted orthoferrites introduce an additional spin moment and an additional orbital moment. The orthoferrites show a spin reorientation transition at temperatures near 80 K, which we can detect by its spin magnetoresistance [17]. At the spin reorientation temperature, the antiferromagnetic modes soften and shift to lower frequencies. Indeed, for a $TmFeO_3$ single crystal, we can observe magnetic excitations in zero field at 73 GHz, that show a nontrivial behavior of the dispersion with the applied magnetic field. They will be investigated in more detail for their potential use in NIMFEIA.

While orbital contributions can be detrimental to the idea to reach the nonlinear excitation regime of magnonic excitations there can be exceptions. We expect CoO to be one of these materials. It is reported to possess a large orbital contribution to its magnetic moment, but both spin and orbital contributions are mainly conserved and hybridize only in a part of the Brillouin zone [18]. Accordingly, Satoh et al. succeeded to excite magnons by all-optical excitation of magnons with frequencies up to 9 THz in antiferromagnetic CoO with an unquenched orbital momentum [19]. They investigated magnon modes that are coupled oscillations of spin and orbital momenta with comparable amplitudes. This provides possibilities to develop magneto-optical devices operating at several terahertz with high output-to-input ratio. We had already worked on CoO thin films by investigating them using Kerr microscopy and photoemission microscopy. For CoO/Pt bilayers we found that we can switch the antiferromagnetic domains with electrical currents. By comparing the results of these electrical measurements to XMLD-PEEM imaging of the antiferromagnetic domain structure before and after the application of current pulses, we revealed the reorientation of the Neel vector in ultra-thin CoO(4 nm). This allowed us to determine that even opposite resistance changes can result from a thermo-magnetoelastic switching mechanism [20]. While Pt offers high spin orbit torques, we found recently that the orbital contributions can be dominant in CoO [21]. In this magnet, which is dominated by orbital angular momentum, we demonstrated a very strong enhancement in orbital Hall magnetoresistance in CoO/Cu*, compared to CoO/Pt. This arises from unique interactions between dynamic orbital angular momentum from surface oxidized Cu* (i.e., the orbital current) and static orbital angular momentum in the antiferromagnetic insulator CoO. Our results show how by using orbital angular momentum-dominated materials, we can harness the benefits of giant orbital currents that have not been possible using conventional spin-dominated magnets, for orbitronics-based devices, offering unprecedented energy efficiency for operations of antiferromagnets that combine ultimate stability with THz dynamics. We further optimize the deposition of CoO in order to fine tune the dynamic properties in cooperation with colleagues from Tohoku university. Figure 4 shows x-ray diffraction of a high-quality CoO thin film, where Laue oscillations next to the film peak indicate a coherent crystal growth from the substrate to the surface. A fit of these Laue oscillations reveals that the strain is relaxing away from the substrate interface.

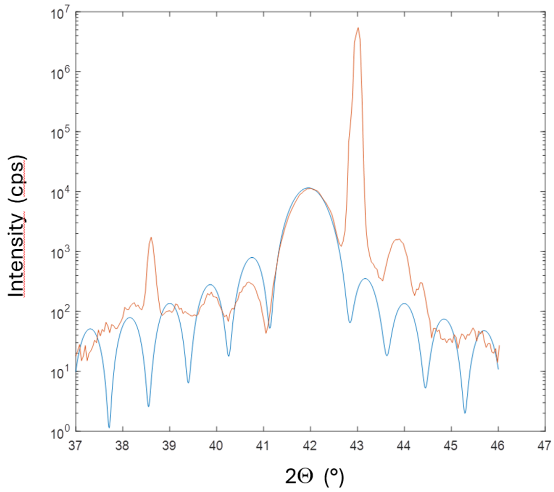


Figure 4: Θ/2Θ x-ray diffraction scan of a CoO film (11 nm) on MgO substrate (red line). The two sharp peaks are from K_{α} and K_{β} diffraction at the substrate peak. A fit with a kinematic scattering model and strain relaxation away from the substrate is shown as a blue line.

1.3. Summary and conclusions

In conclusion, we investigated different oxide materials promising higher frequency dynamics as scheduled for Task 7.2. In experiments, all investigated materials YIG/GIG, NiO, CoO, Fe_2O_3, and $TmFeO_3$ show the expected higher frequency dynamics. The addition of these antiferromagnets to the field of magnonics is just at its beginning. We expect to continue with these material classes, as they promise the most viable way to achieve higher frequency dynamics in future applications. More detailed results than mentioned in this report on YIG/GIG can be found in the scientific paper that is published with NIMFEIA acknowledgements in [A]. Details of the high frequency dynamics of the other materials will be published in future within NIMFEIA.

2. Publications

[A] **Temperature dependent study of the spin dynamics of coupled $Y_3Fe_5O_{12}$/$Gd_3Fe_5O_{12}$/Pt trilayers,** Felix Fuhrmann, Sven Becker, Akashdeep Akashdeep, Gerhard Jakob, Zengyao Ren, Mathias Weiler, and Mathias Kläui,
submitted Phys. Rev. B (2025), https://arxiv.org/pdf/2303.15085,

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