H2020 FET-Open Research and Innovation Actions Project Number 766566
Antiferromagnetic spintronics (ASPIN)

Work package 2, Deliverable D2.1: Report on optical and terahertz experiments

This report summarizes the work of the ASPIN project consortium on optical and THz detection and excitation of dynamics in antiferromagnets. It spans a broad range of works from developing the tools with high temporal and spatial resolution and studies of the fundamentals of spin-dynamics, to demonstrations of THz-pulse electrical switching of antiferromagnetic memory devices and THz spintronic emitters. Apart from the reference to our comprehensive review, covering our as well as world-wide research in the field, the report outlines our original results in selected specific topics. We give references to the corresponding publications featuring details of these results and for each topic we also explicitly list the contributing teams from the consortium comprising: Institute of Physics in Prague (IOP), University of Nottingham (NOT), Max-Planck Institutes (MPG), IGS Ltd. (IGS), Charles University in Prague (CHU), Johannes Gutenberg University in Mainz (JGU).

Contents

1 Review on antiferromagnetic opto-spintronics

Contributing teams: CHU, MPG

Control and detection of spin order in ferromagnetic materials is the main principle enabling magnetic information to be stored and read in current technologies. Antiferromagnetic materials, on the other hand, are far less utilized, despite having some appealing features. For instance, the absence of net magnetization and stray fields eliminates crosstalk between neighbouring devices, and the absence of a primary macroscopic magnetization makes spin manipulation in antiferromagnets inherently faster than in ferromagnets. However, control of spins in antiferromagnets requires exceedingly high magnetic fields, and antiferromagnetic order cannot be detected with conventional magnetometry. In this review [1] we provide an overview and illustrative examples of how electromagnetic radiation can be used for probing (see Fig. 1) and modification of the magnetic order in antiferromagnets. We also discuss possible research directions that are anticipated to be among the main topics defining the future of this rapidly developing field.

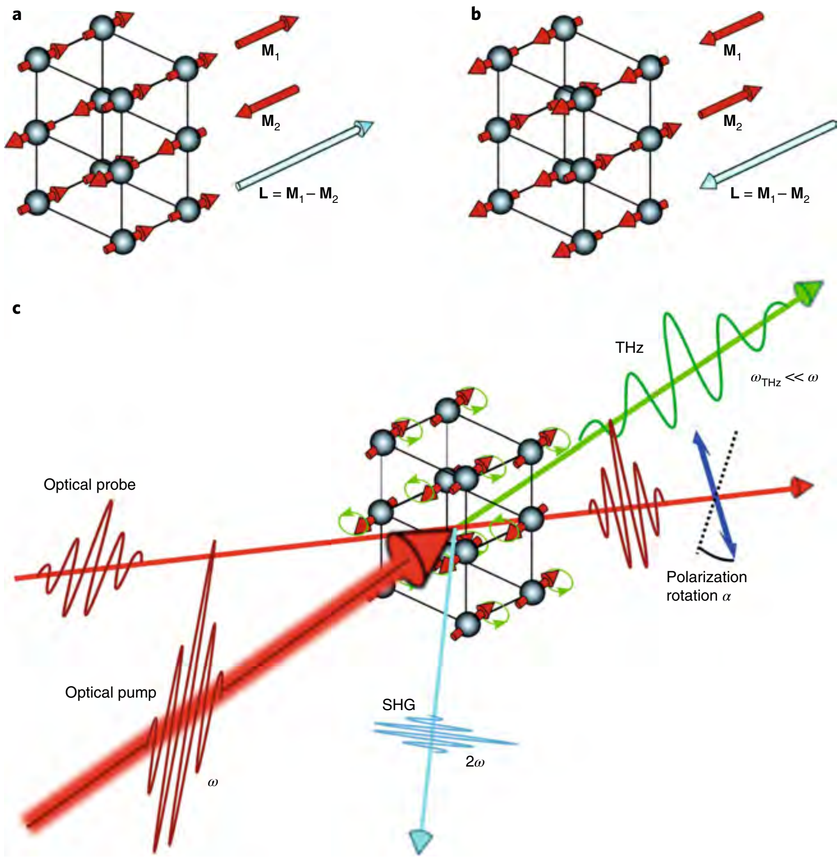

Figure 1: Schematics of investigation of antiferromagnets by electromagnetic radiation [1].

2 Optical detection of antiferromagnetic moments in CuMnAs

2.1 Optical determination of the Néel vector in a CuMnAs thin-film antiferromagnet

Contributing teams: CHU, IOP, NOT, MPG

The absence of a net magnetic moment and ultrashort magnetization dynamics timescales make antiferromagnets notoriously difficult to study using common magnetometers or magnetic resonance techniques. In this work [2] we have demonstrated the experimental determination of the Néel vector in a thin film of antiferromagnetic CuMnAs, a prominent material used in the first realization of antiferromagnetic memory chips. We use a table-top femtosecond pump-probe magneto-optical experiment that is considerably more accessible than the traditionally employed large-scale-facility techniques such as neutron diffraction or X-ray magnetic dichroism measurements.

2.2 Investigation of exchange coupled bilayer Fe/CuMnAs by pump-probe experiment

Contributing teams: CHU, IOP, NOT, MPG

In this work we employed [3] time-resolved pump-probe magneto-optical method to study Fe/CuMnAs bilayer. The probe polarization dependence was used to identify and separate parts of detected signals due to Faraday and Voigt magneto-optical effects that provide information about pump-induced magnetization precession in ferromagnetic Fe and reduction of the sublattice magnetization in antiferromagnetic CuMnAs layers, respectively. We observe a strong asymmetry in the dependence of the precession phase on the external magnetic field that we attribute to the exchange coupling between Fe and CuMnAs. Unlike in X-ray magnetic linear dichroism experiments, we do not observe any significant reorientation of magnetic moments in CuMnAs by external magnetic field due to the interlayer exchange coupling with Fe. Differences between these two experimental techniques, providing the distinct pictures, are discussed.

3 Time-resolved optical detection of laser-pulse induced dynamics in insulating antiferromagnets

3.1 Ultrafast quantum dynamics of the order parameter in antiferromagnets induced by femto-nanomagnons

Contributing teams: JGU, IOP

The impulsive generation of high-energy two-magnon modes in antiferromagnets by femtosecond optical pulses (see Fig. 2), so-called femto-nanomagnons, leads to coherent longitudinal oscillations of the antiferromagnetic order parameter that cannot be described by a thermodynamic Landau-Lifshitz approach. In this work [4] we have formulated a quantum mechanical description in terms of magnon pair operators and coherent states to derive an effective macroscopic equation of motion for the dynamics of the antiferromagnetic order parameter. An implication of the latter is that the photo-induced spin dynamics represents a macroscopic entanglement of pairs of magnons with femtosecond period and nanometer wavelength. By performing magneto-optical pump-probe experiments with 10 femtosecond resolution in the cubic $KNiF_3$ and the uniaxial K_2NiF_4 collinear Heisenberg antiferromagnet, we observed coherent oscillations at the frequency of 22 THz and 16 THz, respectively. The detected frequencies as a function of the temperature ideally fit the two-magnon excitation up to the Néel point. The experimental signals are described as dynamics of magnetic birefringence due to longitudinal oscillations of the antiferromagnetic vector. The oscillations are triggered as a result of laser-induced changes of the exchange interaction.

3.2 Dissecting spin-phonon equilibration in ferrimagnetic insulators by ultrafast lattice excitation

Contributing team: MPG

To gain control over magnetic order on ultrafast time scales, a fundamental understanding of the way electron spins interact with the surrounding crystal lattice is required. However, measurement and analysis even of basic collective processes such as spin-phonon equilibration have remained challenging. In this work [5] we directly probe the flow of energy and angular momentum in the model insulating ferrimagnet yttrium iron garnet (see Fig. 3). After ultrafast resonant lattice excitation, we observe that magnetic order reduces on distinct time scales of 1 ps and 100 ns. Temperature-dependent measurements, a spin-coupling analysis, and simulations show that the two dynamics directly reflect two stages of spinlattice equilibration. On the 1-ps scale, spins and phonons reach quasi-equilibrium in terms of energy through phonon-induced modulation of the exchange interaction. This mechanism leads to identical demagnetization of the ferrimagnet?s two spin sublattices and to a previously inaccessible ferrimagnetic state of increased temperature yet unchanged total magnetization. Finally, on

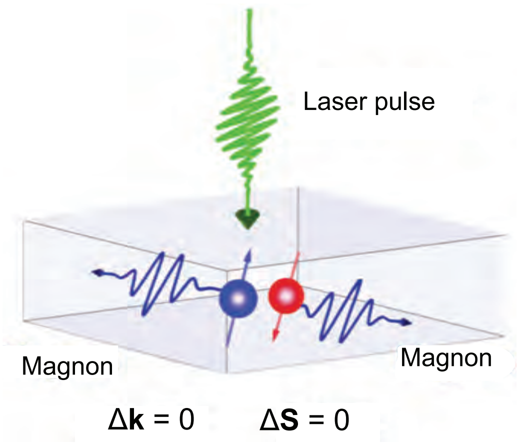

Figure 2: Schematics of photo-induced pairs of magnons via impulsive stimulated Raman scattering [4].

the much slower, 100-ns scale, the excess of spin angular momentum is released to the crystal lattice, resulting in full equilibrium. Our findings are relevant for all insulating ferrimagnets and indicate that spin manipulation by phonons, including the spin Seebeck effect, can be extended to antiferromagnets and into the terahertz frequency range.

4 Magneto-optical microscope integrated in a pump-probe experimental setup

Contributing teams: CHU, IOP, MPG

In this work [6], we describe our experimental setup for a spatially resolved pump-probe experiment with an integrated wide-field magneto-optical (MO) microscope (see Fig. 4). The functionality of the Voigt effect-based microscope was tested using an in-plane magnetized ferromagnetic semiconductor (Ga,Mn)As. It was revealed that the presence of mechanical defects in the (Ga,Mn)As epilayer alters significantly the magnetic anisotropy in their proximity. The importance of MO experiments with simultaneous temporal and spatial resolutions was demonstrated using a (Ga,Mn)As sample attached to a piezoelectric transducer, which produces a voltage-controlled strain. We observed a considerably different behavior in different parts of the sample that enabled us to identify sample parts where the epilayer magnetic anisotropy was significantly modified by the presence of the piezoelectric transducer and where it was not. We discuss the possible applicability of our experimental setup for the research of compensated antiferromagnets, where only MO effects even in magnetic moments are present.

5 Writing by THz pulses in a CuMnAs antiferromagnetic memory

Contributing teams: IOP, MPG, NOT, JGU

The speed of writing of state-of-the-art ferromagnetic memories is physically limited by an intrinsic gigahertz threshold. Our recent realization of memory devices based on antiferromagnets has moved research in an alternative direction. In this work [7] we experimentally demonstrate at room temperature that the speed of reversible electrical writing in a memory device can be scaled up to terahertz using an antiferromagnet (see Fig. 5). A current-induced spin-torque mechanism is responsible for the switching in our memory devices throughout the 12-order-of-magnitude range of writing speeds from hertz to terahertz. Our work opens the path toward the development of memory-logic technology reaching the elusive terahertz band.

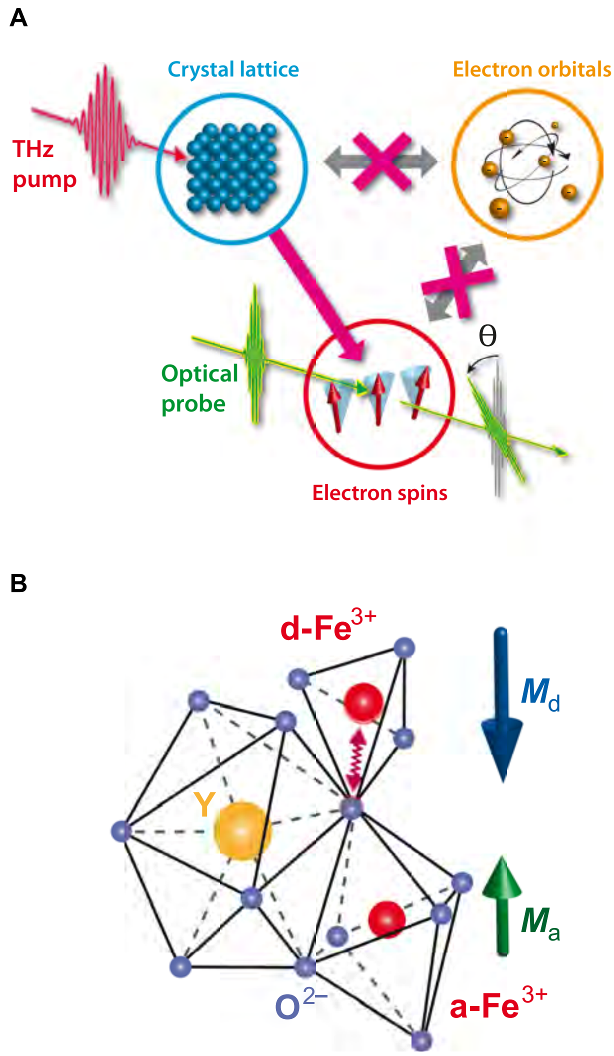

Figure 3: Ultrafast probing of spin-phonon interactions in ferrimagnetic YIG [5].

6 THz spectroscopy, spin-currents, and spintronic emitters

6.1 Femtosecond formation dynamics of the spin Seebeck effect revealed by terahertz spectroscopy

Contributing teams: MPG, JGU

Understanding the transfer of spin angular momentum is essential in modern magnetism research. A model case is the generation of magnons in magnetic insulators by heating an adjacent metal film. In this work [8] we reveal the initial steps of this spin Seebeck effect with 27 fs time resolution using terahertz spectroscopy on bilayers of ferrimagnetic yttrium iron garnet and platinum (see Fig. 6). Upon exciting the metal with an infrared laser pulse, a spin Seebeck current arises on the same 100 fs time scale on which the metal electrons thermalize. This observation highlights that efficient spin transfer critically relies on carrier multiplication and is driven by conduction electrons scattering off the metal-insulator interface. Analytical modeling shows that the electrons? dynamics are almost instantaneously imprinted onto spin current because their spins have a correlation time of only 4 fs and deflect the ferrimagnetic moments without inertia. Applications in material characterization, interface probing, spin-noise spectroscopy and terahertz spin pumping emerge.

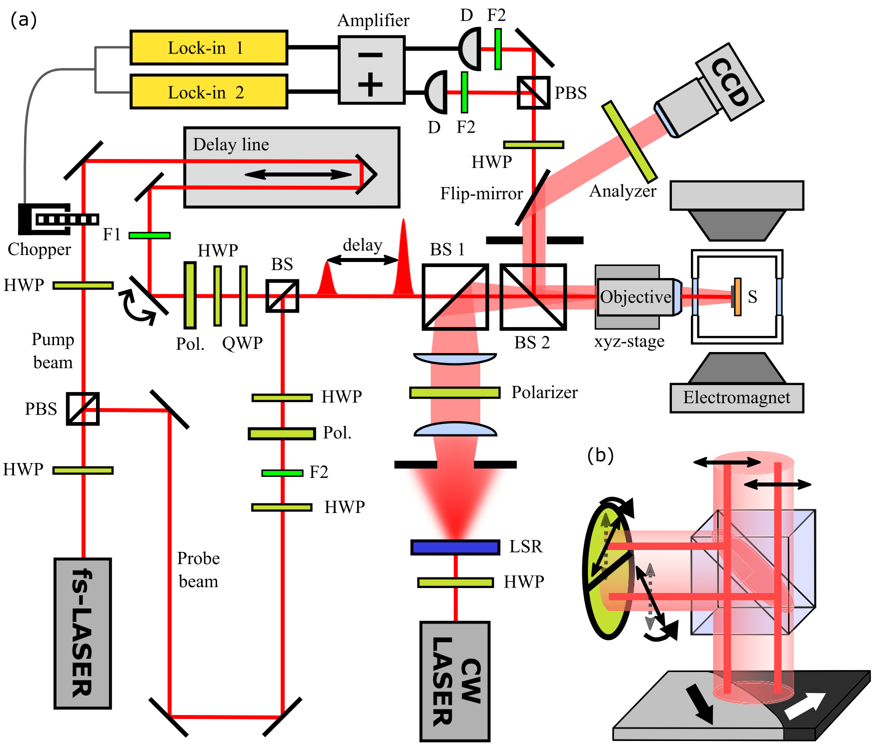

Figure 4: Pump-probe experimental setup with a high spatial resolution combined with a wide-field MO microscope [6].

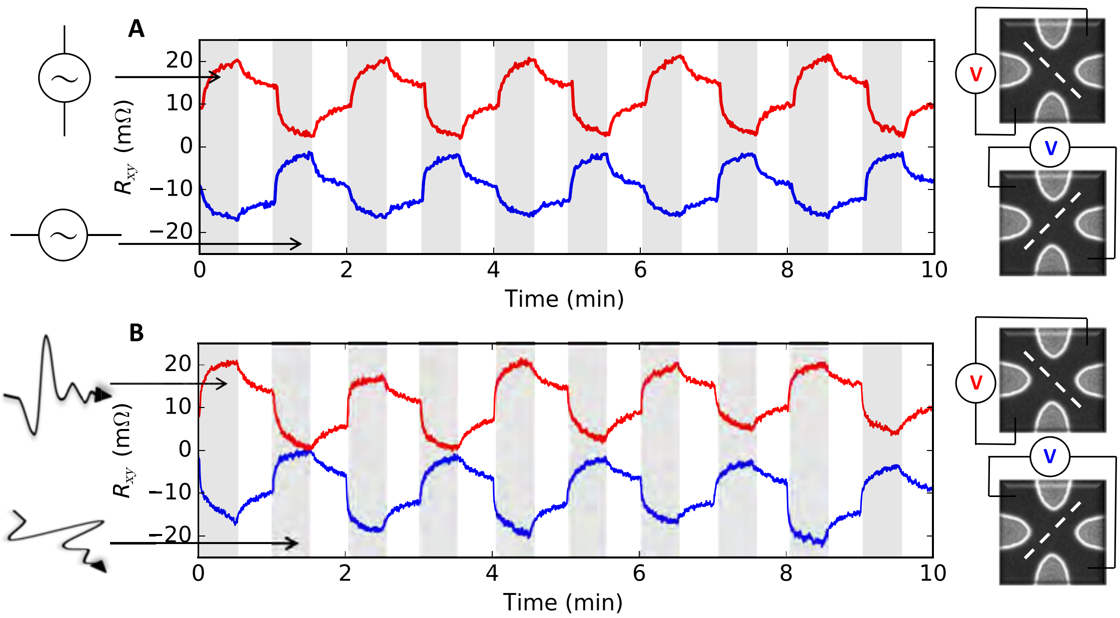

Figure 5: Comparison of switching in a CuMnAs memory cell by microsecond and picosecond pulses [7].

6.2 Terahertz spectroscopy for all-optical spintronic characterization of the spin-Hall-effect metals Pt, W and Cu80Ir20

Contributing teams: MPG, JGU

Identifying materials with an efficient spin-to-charge conversion is crucial for future spintronic

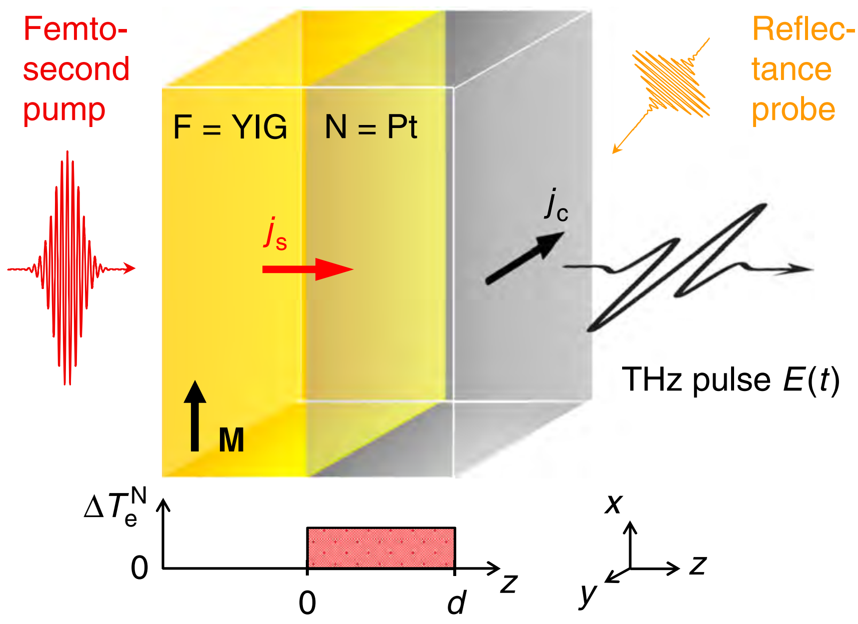

Figure 6: Experimental schematics for probing the ultimate speed of the spin Seebeck effect in a YIG/Pt bilayer [8].

applications. In this respect, the spin Hall effect is a central mechanism as it allows for the interconversion of spin and charge currents. Spintronic material research aims at maximizing its efficiency, quantified by the spin Hall angle and the spin-current relaxation length. In this work [9] we develop an all-optical contact-free method with large sample throughput that allows us to extract these parameters. Employing terahertz spectroscopy and an analytical model, magnetic metallic heterostructures involving Pt, W and Cu80Ir20 are characterized in terms of their optical and spintronic properties. The validity of our analytical model is confirmed by the good agreement with literature DC values. For the samples considered here, we find indications that the interface plays a minor role for the spin-current transmission. Our findings establish terahertz emission spectroscopy as a reliable tool complementing the spintronics workbench.

6.3 Terahertz Spin Currents and Inverse Spin Hall Effect in Thin-Film Heterostructures Containing Complex Magnetic Compounds

Contributing team: MPG

Terahertz emission spectroscopy (TES) of ultrathin multilayers of magnetic and heavy metals has recently attracted much interest. This method not only provides fundamental insights into photoinduced spin transport and spin?orbit interaction at highest frequencies, but has also paved the way for applications such as efficient and ultrabroadband emitters of terahertz (THz) electromagnetic radiation. So far, predominantly standard ferromagnetic materials have been exploited. In this work [10], by introducing a suitable figure of merit, we systematically compare the strength of THz emission from X/Pt bilayers with X being a complex ferro-, ferri- and antiferromagnetic metal, that is, dysprosium cobalt (DyCo5), gadolinium iron (Gd24Fe76), magnetite (Fe3O4) and iron rhodium (FeRh). We find that the performance in terms of spin-current generation not only depends on the spin polarization of the magnet?s conduction electrons, but also on the specific interface conditions, thereby suggesting TES to be a highly interface-sensitive technique. In general, our results are relevant for all applications that rely on the optical generation of ultrafast spin currents in spintronic metallic multilayers.

6.4 Ultrabroadband single-cycle terahertz pulses from a metallic spintronic emitter

Contributing teams: MPG, JGU

In this work [11] we explore the capabilities of metallic spintronic thin-film stacks as a source of intense and broadband terahertz electromagnetic fields (see Fig. 7). For this purpose, we excite a W/CoFeB/Pt trilayer (thickness of 5.6?nm) on a large-area glass substrate (diameter of 7.5?cm) by a femtosecond laser pulse (energy 5.5 mJ, duration 40 fs, and wavelength 800?nm). After focusing, the emitted terahertz pulse is measured to have a duration of 230 fs, a peak field of 300?kV/cm, and an energy of 5 nJ. In particular, the waveform exhibits a gapless spectrum extending from 1 to 10?THz at 10% of its amplitude maximum, thereby facilitating nonlinear control over matter in this difficult-to-reach frequency range on the sub-picosecond time scale.

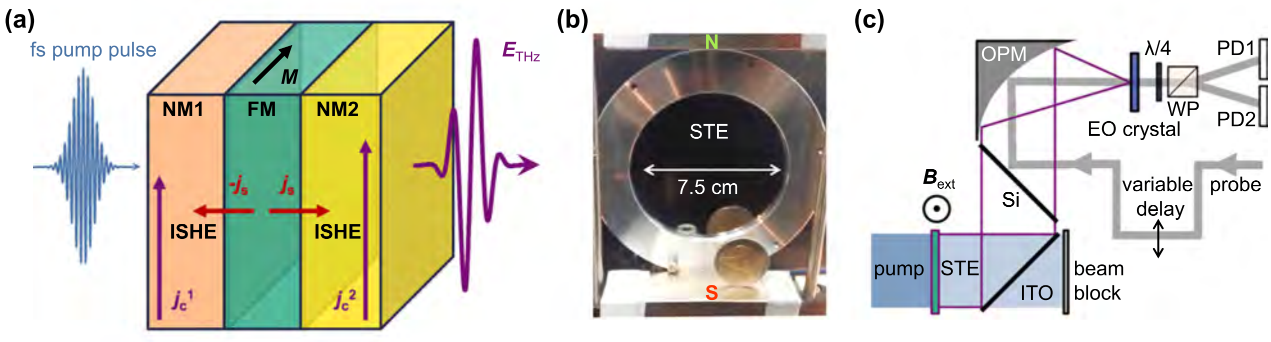

Figure 7: Principle of operation, photograph, and schematic of the experimental setup of a high-field spintronic terahertz emitter [11].

References

[1] Němec, P., Fiebig, M., Kampfrath, T. & Kimel, A. V. Antiferromagnetic opto-spintronics. *Nature Physics* **14**, 229–241 (2018). URL `http://dx.doi.org/10.1038/s41567-018-0051-x`.

[2] Saidl, V. *et al.* Optical determination of the Néel vector in a CuMnAs thin-film antiferromagnet. *Nature Photonics* **11**, 91–96 (2017). `1608.01941`.

[3] Saidl, V. *et al.* Investigation of exchange coupled bilayer Fe/CuMnAs by pump–probe experiment. *Physica Status Solidi - Rapid Research Letters* **11**, 1600441 (2017).

[4] Bossini, D. *et al.* Ultrafast quantum dynamics of the order parameter in antiferromagnets induced by femto-nanomagnons (2017). URL `http://arxiv.org/abs/1710.03143`. `1710.03143`.

[5] Maehrlein, S. F. *et al.* Dissecting spin-phonon equilibration in ferrimagnetic insulators by ultrafast lattice excitation. *Science Advances* **4**, eaar5164 (2018). `1710.02700`.

[6] Janda, T. *et al.* Voigt effect-based wide-field magneto-optical microscope integrated in a pump-probe experimental setup. *Review of Scientific Instruments* **89**, 073703 (2018). URL `http://aip.scitation.org/doi/10.1063/1.5023183`.

[7] Olejník, K. *et al.* Terahertz electrical writing speed in an antiferromagnetic memory. *Science Advances* **4**, eaar3566 (2018). URL `http://advances.sciencemag.org/lookup/doi/10.1126/sciadv.aar3566`. `1711.08444`.

[8] Seifert, T. S. *et al.* Femtosecond formation dynamics of the spin Seebeck effect revealed by terahertz spectroscopy. *Nature Communications* **9**, 2899 (2018). URL `http://dx.doi.org/10.1038/s41467-018-05135-2`. `1709.00768`.

[9] Seifert, T. *et al.* Terahertz spectroscopy for all-optical spintronic characterization of the spin-Hall-effect metals Pt, W and Cu$_{80}$Ir$_{20}$. *Journal of Physics D: Applied Physics* **51**, 364003 (2018). URL http://arxiv.org/abs/1805.02193. 1805.02193.

[10] Seifert, T. *et al.* Terahertz Spin Currents and Inverse Spin Hall Effect in Thin-Film Heterostructures Containing Complex Magnetic Compounds. *Spin* **7**, 1740010 (2017). URL http://www.worldscientific.com/doi/abs/10.1142/S2010324717400100. 1705.11069.

[11] Seifert, T. *et al.* Ultrabroadband single-cycle terahertz pulses with peak fields of 300 kV cm-1from a metallic spintronic emitter. *Applied Physics Letters* **110** (2017). 1703.09970.