

Disentangling spin and charge contributions to the THz emission from ultrathin antiferromagnetic heterostructures

Thomas Metzger¹, Takkashi Kikkawa², Eiji Saitoh², Alexey Kimel¹, Davide Bossini³

¹Institute of Molecules and Materials, Radboud University, Nijmegen, Netherlands

²Department of Applied Physics, The University of Tokyo, Tokyo, Japan

³Department of Physics, Konstanz University, Konstanz, Germany
thomas.metzger@ru.nl

Antiferromagnetic (AF) spintronics is highly promising for fundamental research and future applications. Particularly, the potential of antiferromagnets lies in their robustness against perturbing external electromagnetic fields and their dynamics, intrinsically faster than in ferromagnets, as the magnetic eigenfrequencies enter the terahertz (THz) range [1-2]. The ability to convert the spin dynamics into a charge signal is key to spintronics inspiring recent studies on heavy metal / AF heterostructures [3].

In particular, thin film Pt/NiO heterostructures have become a model system in antiferromagnetic spintronics. Laser-induced THz emission as a result of spin-current injection from NiO [4], optically triggered torque on NiO spins as a consequence of femtosecond laser heating of Pt [5] and magneto-optical pump-probe experiments investigating the ultrafast demagnetization of antiferromagnetic NiO sublattices [6] were reported. This rich variety of the reported and sometimes discrepant effects in the literature sparks controversy and arises from the complexity of these heterostructures rendering the identification of the microscopic origin of the laser-induced THz emission to be challenging.

Here, we suggest that disentangling authentic magnetic from non-magnetic contributions to the THz emission, requires measurements as a function of an external magnetic field intense enough to modify the spin configuration in the ground state. Unprecedented measurements as a function of an external magnetic field up to $\mu H_{ext} = 7$ T allow us to reveal two distinct mechanisms of THz emission from Pt/NiO heterostructures. The first contribution to the emitted THz radiation does not depend on the applied magnetic field, which excludes laser-induced spin dynamics while suggesting optical difference frequency generation. The second contribution is characterized by a linear dependence on the externally applied magnetic field disclosing a magnetic origin. In particular, we interpret this contribution in terms of ultrafast laser-induced quenching of the magnetization, originating from a magnetic field-induced canting of the spin-sublattices in NiO. Our work constitutes an unambiguous experimental approach to access the debated nature

of THz emission observed from heavy metal - antiferromagnetic heterostructures.

References

- [1] Němec, P. et al. opto-spintronics. *Nature Phys* 14, 229–241 (2018)
- [2] Jungwirth, T. et al. Antiferromagnetic spintronics. *Nature Nanotech* 11, 231–241 (2016)
- [3] Kholid, F. et al. The importance of the interface for picosecond spin pumping in antiferromagnet-heavy metal heterostructures. *Nat Commun* 14, 538 (2023)
- [4] Qiu, H., Zhou, L., Zhang, C. et al. Ultrafast spin current generated from an antiferromagnet. *Nat. Phys.* 17, 388–394 (2021)
- [5] Rongione, E. et al. Emission of coherent THz magnons in an antiferromagnetic insulator triggered by ultrafast spin–phonon interactions. *Nat Commun* 14, 1818 (2023)
- [6] Wust S. et al. Indirect optical manipulation of the antiferromagnetic order of insulating NiO by ultrafast interfacial energy transfer arXiv:2205.02686 (2022)