Origins of THz radiation from ultrafast light-driven ferromagnets and antiferromagnetic Mott insulators

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The ultrafast demagnetization is a surprising phenomenon discovered in 1996 in which a femtosecond laser pulse (fsLP) pumping of a ferromagnet leads to its demagnetization, as nonclassical dynamics where magnetization vector is shrinking in length while not rotating. Despite being one of the primary experimental observables, THz electromagnetic radiation from either a single magnetic layer, or much more enhanced from ferromagnet/spin-orbit-material bilayers [1], has rarely been calculated. This has forced experimentalists to develop intuitive pictures [1], such as of interlayer spin current generated by "spin voltage" that is converted into charge currents via spin-orbit coupling induced mechanisms. This talk will discuss the recently developed framework [2], combining time-dependent density functional theory (TDDFT) and Jefimenko formulas for time-retarded electric and magnetic fields as proper solutions of the Maxwell equations, that make it possible to compute emitted THz radiation from first principles. Inspection of time-dependent charge currents, as the only source of the electric field in the far-field region within Jefimenko formulas, then reveals that a major contribution to THz radiation arises from *overlooked* intralayer charge current within the ferromagnetic metallic layer that is pumped by demagnetization dynamics. Interlayer spin current is also pumped by ultrafast demagnetization dynamics [3], flowing toward spin-orbit-material layer where it is converted into charge current as often assumed. Its origin can be explained [4] by the same time-dependent quantum transport calculations which account for conventional spin pumping by rotating magnetization of fixed length when ferromagnets are driven by microwave photons of much lower energy than in ultrafast demagnetization phenomena. Finally, in the case of fsLP-driven antiferromagnetic Mott insulators, as motivated by experiments [5] on NiO/Pt bilayers, standard TDDFT is not applicable due to intricacies of handling time-dependent Hubbard U. We replace it with tensor network and/or exact diagonalization calculations for multi-orbital Hubbard-Hund-Heisenberg model which includes proximity spin-orbit coupling due to adjacent heavy metal Pt layer. This reveals [6] nonclassical dynamics of the Néel vector and nonequilibrium magnetization, which leads to both THz radiation and peculiar high harmonic generation, including the *noninteger* ones, while *not requiring* any interlayer spin transport as in the case of ferromagents as also confirmed experimentally [5].

References

[1] T. S. Seifert, D. Go, H. Hayashi, R. Rouzegar, F. Freimuth, K. Ando, Y. Mokrousov, and T. Kampfrath, Nat. Nanotechnol. **18**, 1132 (2023).
[2] A. Kefayati and B. K. Nikolić, Phys. Rev. Lett. **133**, 136704 (2024).
[3] A. Kefayati, Y. Ren, M. B. Jungfleisch, L. Gundlach, J. Q. Xiao, and B. K. Nikolić, Phys. Rev. B **111**, L140415 (2025).
[4] J. Varela-Manjarres, A. Kefayati, M. B. Jungfleisch, J. Q. Xiao, and B. K. Nikolić, Phys. Rev. B **110**, L060410 (2024) (accompanied by popular highlight at https://physics.aps.org/articles/v17/s97).
[5] T.W. J. Metzger *et al.*, Phys. Rev. Lett. **135**, 076702 (2025).
[6] F. Garcia-Gaitan, A. E. Feiguin, and B. K. Nikolić, Phy s. Rev. Lett. **135**, 086704 (2025).

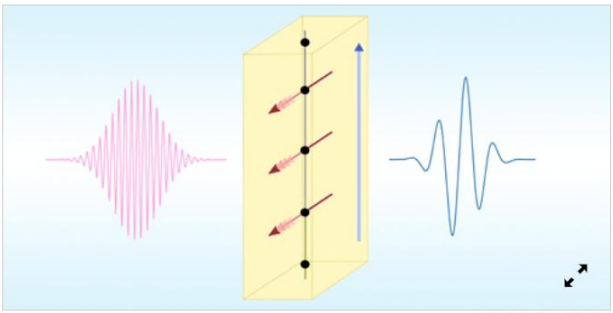

Fig. 1. For the first time in 20 years (since the first observation of THz radiation by demagnetizing in 2004 by Beaurepaire et al.) we have demonstrated how to microscopically compute THz radiation from single ferromagnetic [2,3,4] or antiferromagnetic layer [6], as well as their bilayers [2,3,4,6] with heavy metals. This approach has unearthed new mechanisms behind such emission, such as charge pumping by magnetization which is changing length without rotation due to ultrafast light