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Optical control of 4f orbital state in rare-earth metals

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Ultrafast magnetic response to optical excitation has been studied for many years; still, only very recently it has been appreciated that early electronic excitations may play a more important role than just being a transient step in the deposition of energy in the electron system [1,2]. An example is the optically induced spin transfer (OISTR) in 3d materials where the excited 3d electrons together with band structure effects directly lead to spin transfer processes. In the case of 4f metals, f-f electronic excitations within the 4f shell are dipole forbidden. Therefore, a variation of the 4f electronic structure was generally assumed to be negligible for the ultrafast 4f magnetic response to optical excitations. From pioneering experiments at the European XFEL [3] and FLASH where we combine time-resolved X-ray absorption and resonant inelastic X-ray scattering (RIXS), we learned that after exciting the 5d6s valence electrons in Tb metal with 800 nm laser pulses, the 4f electronic state actually can be affected on ultrashort time scales. The 4f electronic excitations are mainly driven by 5d-4f electron-electron scattering, but we also find indications of 5d-4f electron transfer contributing to the observed dynamics. The spin and orbital momenta derived from the 4f electronic state define the coupling of the magnetic system to the environment. We observe 20% of the Tb atoms with an altered orbital state, that results in a change of the magneto crystalline anisotropy. Our study gives a new dimension to the discussion about optically control of 4f magnetic dynamics, as it provides a femtosecond handle on the coupling between 4f magnetic system and environment.

[1] F. Willems et al., Optical inter-site spin transfer probed by energy and spin-resolved transient absorption spectroscopy, Nat. Commun. 11, 871 (2020).
[2] E. Golias et al., Ultrafast Optically Induced Ferromagnetic State in an Elemental Antiferromagnet Phys Rev Lett 126, 107202 (2021).
[3] N. Thielemann-Kühn et. al., Optical control of 4f orbital state in rare-earth metals, (2021) (https://arxiv.org/abs/2106.09999)

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