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Controlling magnetism with light in a zero orbital angular momentum antiferromagnet

Mattias Matthiesen,[1,5] Jorrit Hortensius,[1] Samuel Manas-Valero,[2] Makars Siskins,[1] Boris Ivanov,[3] Herre van der Zant,[1] Eugenio Coronado,[2] Dmytro Afanasiev,[4] and Andrea Caviglia[1,5]

[1] *Kavli Institute of Nanoscience, Delft University of Technology, P.O. Box 5046, 2600 GA Delft, Netherlands*
[2] *Instituto de Ciencia Molecular (ICMol), Universitat de Valencia, Catedrático José Beltrán 2, 46980 Paterna, Spain.*
[3] *Institute of Magnetism, National Academy of Sciences and Ministry of Education and Science, 03142 Kyiv, Ukraine*
[4] *Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, Netherlands*
[5] *DQMP-University of Geneva, École de Physique, 24, Quai Ernest-Ansermet, CH-1211 Geneva, Switzerland*

Antiferromagnetic materials feature intrinsic ultrafast spin dynamics, making them ideal candidates for future magnonic devices operating at THz frequencies. A major focus of current research is the investigation of optical methods for the efficient generation of coherent magnons in antiferromagnetic insulators. In magnetic lattices endowed with orbital angular momentum, spin-orbit coupling enables spin dynamics through the resonant excitation of low-energy electric dipoles such as phonons and orbital resonances which interact with spins. However, in magnetic systems with zero orbital angular momentum, microscopic pathways for the resonant and low-energy optical excitation of coherent spin dynamics are lacking. Here, we consider experimentally the relative merits of electronic and vibrational excitations for the optical control of zero orbital angular momentum magnets, focusing on a limit case: the antiferromagnet manganese thiophoshate (MnPS3), constituted by orbital singlet Mn2+ ions. We study the correlation of spins with two types of excitations within its band gap: a bound electron orbital excitation from the singlet orbital ground state of Mn2+ into an orbital triplet state, which causes coherent spin precession, and a vibrational excitation of the crystal field that causes thermal spin disorder. Our findings cast orbital transitions as key targets for magnetic control in insulators constituted by magnetic centers of zero orbital angular momentum.