

## Poster-1-30

## Applying Coherent Two-Dimensional Spectroscopy to a Pulsed Magnetophononic System

Baptiste Demazure<sup>1,2</sup>, Götz Uhrig,<sup>3</sup> and Bruce Normand<sup>1,2</sup>

<sup>1</sup> LTC, Paul Scherrer Institute, 5232 Villigen-PSI, Switzerland

<sup>2</sup> Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

<sup>3</sup> 3TU Dortmund University, Otto-Hahn-Straße 4, 44221 Dortmund, Germany

Coherent two-dimensional spectroscopy is a powerful tool to distinguish non-linear contributions to the non-equilibrium physics of quantum systems. The technique has no frequency-range limitation and thus has recently been used in the terahertz regime to observe the high-order response of magnon excitations [1]. In linear magnetophononics, the modulation of magnetic interactions by laser-driven phonons, the coupled phononic and magnetic excitations both have frequencies similar to that of the driving pulse. In this work we use the new frequency axis offered by coherent two-dimensional spectroscopy to isolate the non-linear response of such a system by applying the methodology to a straightforward model of a gapped quantum spin chain with strong magnetophononic coupling [2]. Pulsed driving in this model reveals non-linear contributions of hybrid phononic and magnetic nature, including the sum and difference frequencies of composite phonon-bitriplon excitations [3]. We demonstrate that coherent two-dimensional spectroscopy offers both qualitative and quantitative separation of the different non-linear contributions emerging at strong coupling in pulsed magnetophononics.

[1] J. Lu, X. Li, H. Y. Hwang, B. K. Ofori-Okai, T. Kurihara, T. Suemoto, and K. A. Nelson, Coherent Two-Dimensional Terahertz Magnetic Resonance Spectroscopy of Collective Spin Waves, *Phys. Rev. Lett.* 118, 207204 (2017).

[2] M. Yarmohammadi, M. Krebs, G. S. Uhrig and B. Normand, Strong-coupling magnetophononics: Self-blocking, phonon-bitriplons, and spin-band engineering, *Phys. Rev. B* 107, 174415 (2023).

[3] B. Demazure, M. Krebs, G.S. Uhrig and B. Normand, Pulsed magnetophononics in a gapped quantum spin chain, unpublished (2024).