Signature of Jahn-Teller-like magnetic instability in $YbFeO_3$ and dynamic strong coupling of Fe spins and Yb orbitals

O. Y. Kovalenko[a], N. R. Vovk[a], A. Wu[b], R. V. Mikhaylovskiy[a]
[a] *Lancaster University, United Kingdom;*
[b] *Shanghai Institute of Ceramics, China.*

Magnetic recording is a central technology for long-term data storage. Understanding the physics of ultrafast angular momentum transfer between light and spins is one of the key problems in magnetism. Light-driven magnetisation control may lead to revolutionary all-optical magnetic recording technology. Antiferromagnetic materials, such as $YbFeO_3$, have emerged as promising candidates for future storage media [1,2]. They offer unique functionalities, such as ultrafast terahertz spin dynamics and low-energy excitation, thanks to the direct interaction of the THz pulse with a spin system [3].

Ultrashort terahertz (THz) pulses were used to excite low-energy orbital transitions in $YbFeO_3$ to reveal angular momentum dynamics at high excitation strengths. The magnetisation in this material arises mainly from the spins of the 3d ions, whereas the interaction of the spins with the 4f electronic orbitals sets the character of the magnetic configuration. The $YbFeO_3$ exhibits a spin reorientation transition (SRT) of the Jahn-Teller type at temperatures below 9 K. This SRT originates from the interplay between two anisotropy energies: the Yb-Fe exchange and crystalline anisotropy. The SRT involves rotations of both Iron spins and Ytterbium orbital moment. These unique properties of $YbFeO_3$, including its energy anti-crossing, make it a vibrant playground for THz applications. Despite this unique phase diagram, the ultrafast dynamics of YbFeO3 have not yet been studied.

We explored the dynamics in $YbFeO_3$ using a time-resolved pump-probe technique across the SRT. Fig. 1 depicts the Fourier spectral map of the measured polarisation rotation, shown as a function of the sample temperature for the three combinations of the sample orientation and detection conditions. Four well-distinguished modes were identified as: 1 – quasi-antiferromagnetic (q-AFM), 2 – quasi-ferromagnetic (q-FM), 3,4 – two orbital rare-earth modes (R_1, R_2). While the q-AFM and R_1 are weakly interacting and show only a small frequency shift, the q-FM and the low-lying R_2 are strongly coupled, which is reflected in the kink-like behaviour of the q-FM and softening of the R_2 mode. The origin and properties of this interaction will be discussed, along with a theoretical description that yields good agreement with experimental results (red curves on Fig. 1). We believe that our findings represent a major step in understanding quantum effects and dynamical driving of phase transitions in ultrafast magnetism.

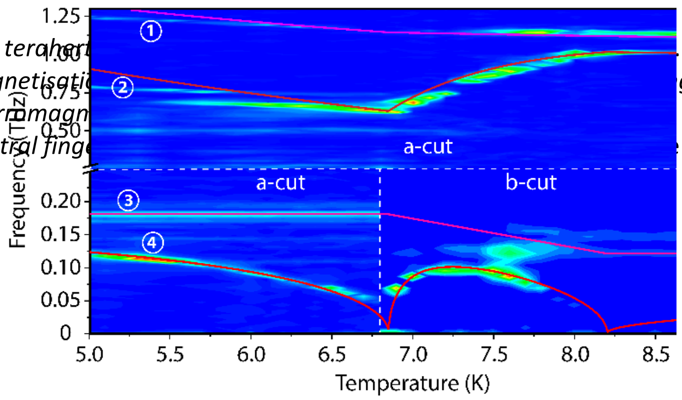

Fig. 1. The map plot of the Fourier spectra of transient Faraday rotation in $YbFeO_3$ as a function of the sample temperature. The four presented modes were identified as 1: q - AFM, 2: q - FM, 3,4: two rare-earth modes. The solid lines are show the theoretically calculated frequencies.

[1] S. Baierl, et al. *Nonlinear spin control by terahert*
[2] R. V. Mikhaylovskiy, et al. *Terahertz magnetisati* *g of Dy^{3+} subsystem in the multisublattice antiferromagn*
[3] S. Schlauderer, et al. *Temporal and spectral finge* **569**, 383 (2019).