# Controlling coherent energy flow between collective THz excitations in condensed matter

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**Abstract:** Coherent control over the magnetization state of the rare-earth orthoferrite $ErFeO_3$ is achieved by nonlinear lattice excitation via mid-infrared laser pulses. This low-dissipative approach enables new pathways in light-driven manipulation of magnetic materials.
**OCIS codes:** (300.6530); (320.7130)

## 1. Introduction and Technical aspects

Direct excitation at THz and mid-infrared frequencies makes it possible to coherently manipulate collective modes of condensed matter. This approach involves the stimulation of low-energy resonances (in the meV energy range) representing therefore a promising route for high-speed and low-dissipation all-optical control of materials.

In this spirit, the control of the crystal lattice at mid-infrared wavelengths has emerged as a new mean to solids manipulation. Recent examples include insulator-metal and magnetic transitions in manganites [1-3] and nickelates [4], as well as the enhancement of superconductivity in cuprates [5]. These results are interpreted, in the framework of nonlinear phononics [6,7], as an energy flow between different modes driven by lattice anharmonicities. A strongly driven infrared-active vibrational mode can couple to a lower frequency Raman mode. Moreover, in a process very similar to optical rectification, the IR-active phonon field, rectified, acts as a directional force that causes an abrupt deformation of the lattice along the coordinate of the Raman excitation [6,7], leading to new collective electronic properties [8].

Beyond nonlinear phononics, the Dzyaloshinskii-Moriya (DM) interaction [9,10] is an anisotropic magnetic interaction that in contrast to isotropic superexchange favors non-collinear ordering of the spins. This interaction develops between two magnetic ions separated by an oxygen ion (Fig.1b). The relative positions of the ions strongly influence the interaction strength making it extremely sensitive to any modification of bond angles and lengths. Therefore, the ability of transiently controlling the lattice may result in the modification of the magnetic ordering in materials characterized by the DM interaction.

To explore this possibility, we performed a mid-IR pump/near-IR probe experiment on the rare-earth orthoferrite, $ErFeO_3$. This material crystallizes in an orthorombically distorted perovskite structure (Fig. 1(a)) [11]. Owing to the DM interaction the antiferromagnetic order is perturbed resulting in a small canting ($< 1°$) of the iron spins along the *c* axis. By pumping resonantly a high-frequency vibrational mode, we show that coherent magnons can be triggered. The coupling of the lattice distortion to the spin system is hypothesized to be driven by the transient modification of the DM interaction.

Carrier-envelope-phase stable 180-fs mid-IR pump pulses were generated by difference-frequency mixing of two optical parametric amplifier outputs. The center frequency was tuned at 20 THz, resonant with a high-frequency $B_u$ symmetry stretching mode. The beam was focused onto the sample resulting in a 5.8 mJ/cm$^2$ excitation fluence. The sample was c-oriented (*c* axis parallel to the wave vector of the pump light and face of the sample parallel to the *ab* plane). Given the giant magneto-optical susceptibility of rare-earth orthoferrites [12], in order to probe the transient modification of the sample magnetization along the *c* axis we exploited the Faraday effect. This phenomenon consists in the rotation of the polarization of a light field travelling through a magnetized medium. Such rotation is directly proportional to the projection of the magnetization of the material along the light propagation direction.

## 2. Results and Discussion

The transient Faraday rotation induced by the mid-IR pump pulse (for a sample temperature of 100 K) is shown in Fig. 1c (upper panel). Clear oscillations at 0.750 THz (long-lived) and 3.35 THz are observed (see also the Fourier transform in Fig. 1d (upper panel)). These frequencies are in excellent agreement with those of the quasi-antiferromagnetic magnon [11] and the $A_{1g}(1)$ Raman phonon [13] of this material, respectively.

The magnon is characterized by a precessional motion of the two spin sublattices that modifies the net magnetization along the *c* axis (Fig. 1d (upper panel)). The coherent phonon oscillations, as mentioned before, are launched by the nonlinearly driven, displacive lattice distortion. The lattice motion associated with the Raman mode involves a stretching of the oxygen atoms in the $FeO_6$ octahedra (depicted in Fig.1d (upper panel)) that leaves the Fe ions unperturbed. Therefore this distortion modifies the relative positions of the Fe and O ions, possibly inducing the change in the anisotropic DM interaction that coherently drives the magnon.

The same measurement was also performed at lower temperatures (30 K), below the spin reorientation transition [13], in which the net magnetization of the material rotates from the *c* axis to the *a* axis. Given our experimental geometry we were then insensitive to any variation of the magnetization, as confirmed by the data (Fig. 1c and 1d (lower panels)). Indeed, in the time-domain trace and its Fourier transform only the oscillations related to the Raman phonon are present, corroborating our physical picture.

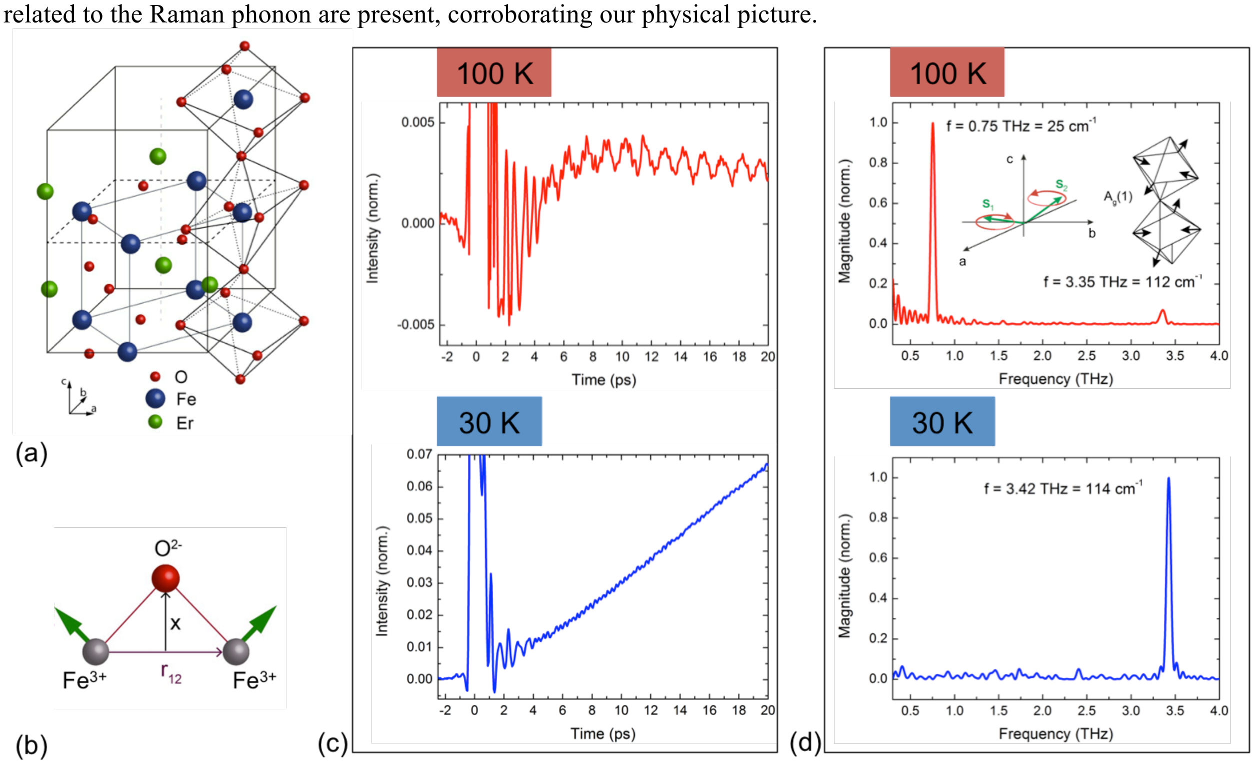


**Figure 1: (a)** Crystal structure of $ErFeO_3$. **(b)** The inset depicts the anisotropic DM interaction between the two Fe spins (green arrows) mediated by the oxygen ion (open circle). The coupling causes canting of the otherwise antiferromagnetically aligned spins, resulting in a ferromagnetic component (here along *x*). **(c)** Transient optical birefringence measured at 100 K (upper panel) and 30 K (lower panel). **(d)** Fourier transform of the oscillatory signal contributions of panel (c), together with the spin and atomic motions of the *quasi-antiferromagnetic* magnon and of the $A_{1g}(1)$ Raman mode, respectively.

Our experiment reports on the capability of exciting coherent magnons via phonon pumping in a rare-earth orthoferrite. The approach presented here can be extended to the magnetic control of more complex materials, also characterized by the DM interaction, such as, e.g. multiferroic perovskites [14].

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