THz-driven coupled dynamics of 4*f* orbitals and 3*d* spins in rare-earth orthoferrites

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The rare-earth orthoferrites (REO) have been attracting significant attention since their discovery in the 1940s due to unique magnetic properties, such as spin reorientation phase transitions (SRPT), strong magneto-optical effects, THz-frequency spin dynamics, multiferroicity, and quantum magnonics. Recent advancements in the development of THz time-domain spectroscopy (THz-TDS) techniques allowed the study of various types of excitations with meV energies, including spin dynamics, by directly interacting with atomic spins using THz radiation. Regarding the REOs, an interesting idea was to use intense THz fields to achieve control over nonlinear SRPT and switch of iron spins due to the changing crystalline magnetic anisotropy via the REO subsystem, driven by THz pulses [1–2]. This raises the question regarding the exchange interaction between rare-earth and iron orbitals in a non-equilibrium state. Moreover, different scenarios for dynamic coupling are possible depending on the type of a rare-earth ion.

To address this issue, we have formulated a theoretical model based on the microscopic approach [3] to elucidate the magnetic switching in REOs with non-Kramers ions subjected to strong THz excitation [1–2]. Using the archetypical orthoferrite $TmFeO_3$ as a model system, we investigated the static properties of the R and Fe subsystems across the SRPT. Employing an adiabatic approximation, we determined the resonance frequencies for the Fe and R magnetic sub-lattices as a function of temperature, aligning our findings with experimental data from [1]. We then performed numerical modelling to accurately describe the behaviour of its anisotropy functions vs temperature. Through numerical modelling, we described the behaviour of anisotropy functions relative to temperature, identified threshold fields for spin switching—whether via Zeeman mechanisms or anisotropy-driven torques altered through the R-subsystem—and estimated the energy dissipation during the switching process, achieving excellent correlation with the experimental values from [2].

Furthermore, we extended our model to interpret our recent experimental results on $TbFeO_3$, a unique member of the orthoferrite family. In our experimental work we employed ultrashort terahertz (THz) pulses to excite low-energy excitations of Fe^{3+} spins and Tb^{3+} atomic-like transitions to reveal the character of their coupling and hybridization. The magnetisation in $TbFeO_3$ mainly arises from the spins of the Fe^{3+} ions, whereas the interaction of the spins with the Tb^{3+} electronic orbitals sets the character of the magnetic configuration. The $TbFeO_3$ exhibits a second-order magnetic phase transition of the Jahn-Teller type at temperatures below 8.5 K. This SRPT originates from the interplay between two anisotropy energies: Tb-Fe exchange and crystalline anisotropy energies. As a result, $TbFeO_3$ exhibits a cross-over (i.e. equality) of the frequencies of the antiferromagnetic resonance mode and atomic-like mode of Tb, which leads to their dynamical repulsion and avoided crossing effect. The coupling is so strong that the frequency of the low-lying hybrid mode is pushed to zero. Using our model, we managed to model dynamics between interacting Fe and Tb magnetic subsystems, and to fit it with an excellent match to our experimental findings (Fig.1).

References

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Figures

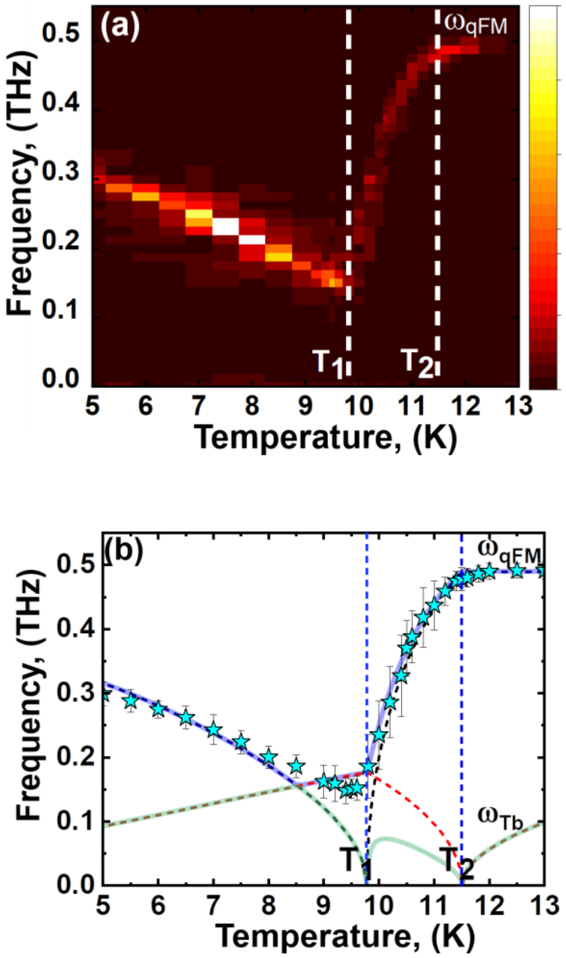

Figure 1. Temperature dependence of the resonance frequencies ω_{qFM} and ω_{Tb} in $TbFeO_3$: panel (a) the Fourier map plot of resonance frequencies obtained from polarisation rotation measurements; panel (b) comparison between experimental data and theoretical curves, accounting for the dynamical repulsion. The dashed lines indicate phase transitions temperaturas T_1 and T_2.