

ABSTRACT

Coherent and deterministic control of magnetization at ultrafast timescales is essential for the development of next-generation spintronic and magnonic technologies. Ferrimagnetic systems offer a unique platform for such studies, as they combine antiferromagnetic-like exchange dynamics in the THz regime with a finite macroscopic magnetization, facilitating efficient excitation and detection of the exchange mode. In this context, terahertz (THz) light fields are ideal for both probing and driving these dynamics. In particular, their magnetic field component couples directly to spins via the Zeeman interaction, while their ultrashort temporal profile enables access to intrinsic exchange-driven timescales without significant thermal effects.

We report the first experimental study of nonlinear THz-driven magnetization dynamics in the Bi-doped rare-earth iron garnet ($\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$). By combining time-domain THz spectroscopy with Faraday probing, we isolated the 0.48-THz Kaplan-Kittel exchange mode. Our results reveal that conventional Zeeman torque models fail to describe the nonlinear dynamics under two-pulse excitation. We propose that a relativistic field-derivative torque (FDT), rooted in spin-orbit coupling, is the critical mechanism required to accurately model these nonlinear responses. Building on this, we have developed a theoretical framework to systematically scrutinize the role of material properties in the FDT-induced THz-driven magnetization dynamics. By incorporating the FDT term into the Landau-Lifshitz-Gilbert equation for a two-sublattice ferrimagnet, we demonstrate that the FDT significantly enhances the amplitude of coherent magnon excitation and induces a phase shift in the ensuing dynamics. Furthermore, we show that material properties like sublattice-dependent Gilbert damping, the gyromagnetic ratio, and the volume-per-spin play a crucial role in determining the distinct nonlinear signals observed under dual-THz excitation.

Finally, we have extended our investigation to temperature-dependent magnetization dynamics near the magnetization compensation point in the same garnet system. We observe the emergence of additional excitations at THz frequencies originating from the crystal electric field (CEF) splitting of the Yb^{3+} ions. These localized CEF modes hybridize with the collective exchange dynamics, leading to a spectral weight redistribution and the appearance of a CEF-mediated exchange mode. This interplay highlights the importance

of orbital degrees of freedom and facilitates the necessity of incorporating multi-sublattice descriptions in the magnetization dynamics, particularly in the vicinity of the compensation temperature.

Overall, the thesis establishes a unified understanding of the ultrafast magnetization dynamics in a ferrimagnetic system by combining nonlinear THz spectroscopy and temperature-dependent multi-sublattice interactions. The results provide fundamental insights into exchange-driven magnetization dynamics and open new pathways for controlling magnetic order at ultrafast timescales.