

Magnetization Dynamics of Antiferromagnetically Coupled Spins at Ultrafast Timescales

By

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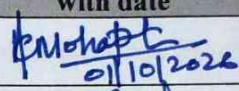
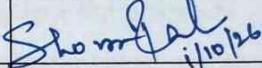
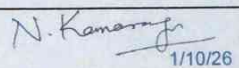
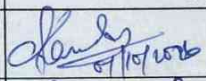
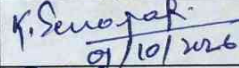


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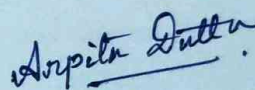
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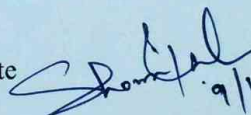
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2. "Role of material-dependent properties in THz field-derivative-torque-induced nonlinear magnetization dynamics", **Arpita Dutta**, Pratyay Mukherjee, Swosti P. Sarangi, Somasree Bhattacharjee, Shovon Pal and Ritwik Mondal, Phys. Rev. Materials, **2024**, 8, 114404.
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2. "Intensity-driven Mott-like transition in a 2D Ruddlesden-Popper perovskite", Koushik Mondal, Amit Haldar, Indrajit Mondal, **Arpita Dutta**, Himanshu Bhatt, Biswajit Mahata, Soumili Banerjee, Hirendra Nath Ghosh, Satyaprasad P Senanayak, and Shovon Pal, (Manuscript accepted in Small).
3. "Field-derivative torque induced magnetization reversal in ferrimagnetic $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ ", Pratyay Mukherjee, **Arpita Dutta**, Shovon Pal and Ritwik Mondal, Phys. Rev. Materials, **2025**, 9, 104405.
4. "2D-THz spectroscopy: Exploring the nonlinear dynamics in quantum materials", **Arpita Dutta**, Payel Shee, Amit Haldar, and Shovon Pal, J. Phys.: Condens. Matter, **2025**, 37, 203002.
5. "Charge carrier dynamics in bandgap modulated covellite-CuS nanostructures", Kusuma Jagadish, Akshath Godha, Bidhan Pandit, Yogesh Jadhav, **Arpita Dutta**, Jyotiprakash Satapathy, Himangshu Bhatt, Balpartap Singh, Surendra Kumar Makineni, Shovon Pal and Sachin R. Rondiya, Small, **2024**, 20, 2405859.
6. "Terahertz conductivity of free-standing 3D covalent organic framework membranes fabricated via triple-layer-dual interfacial approach", Adithyan Puthukkudi, Satyapriya Nath, Payel Shee, **Arpita Dutta**, Chetan V. Rajput, Suresh Bommakanti, Jeevanjyoti Mohaptra, Mahalaxmi Samal, Sarmistha Anwar, Shovon Pal and Bishnu P. Biswal, Adv. Mater., **2024**, 36, 2312960.

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This thesis is dedicated to my beloved parents.

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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.

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Chapter 1

Introduction

The discovery of ultrafast laser-induced demagnetization in ferromagnetic nickel by Beaurepaire et al. in 1996 marked a turning point in modern condensed matter physics, fundamentally changing our understanding of magnetization dynamics far from equilibrium [2]. For the first time, it became possible to probe the evolution of spins on femtosecond timescales and to explore their coupling to charge, lattice, and orbital degrees of freedom. This breakthrough laid the foundation for spintronics and magnonics – fields in which information is carried and processed via spin currents and collective spin excitations [3–7]. In parallel, rapid advances in ultrafast laser and terahertz (THz) technologies have enabled coherent spin manipulation on femtosecond to picosecond timescales, opening a direct route to high-speed, energy-efficient magnetic control [8–12]. Understanding the *linear* and *nonlinear* response of magnetic systems to intense THz fields, particularly in antiferromagnetically coupled systems, where two or more spin sublattices introduce rich internal dynamics – has consequently emerged as a key challenge in contemporary magnetism research, and forms the basis of this thesis.

Magnetically ordered systems are broadly classified into ferromagnets and antiferromagnets according to the relative orientation of neighboring spins. In ferromagnets, spins align parallel to each other, resulting in a finite macroscopic magnetization. Antiferromagnets, in contrast, exhibit antiparallel spin ordering with vanishing net magnetization in equilibrium. While ferromagnets are comparatively easier to manipulate and detect owing to their finite magnetic moment, their dynamical response is generally confined to the gigahertz (GHz) frequency regime [13–15]. Antiferromagnets support ultrafast spin dynamics

in the THz range due to the strong exchange interaction between oppositely aligned sublattices; however, the absence of net magnetization makes the excitation and detection of their collective dynamics experimentally challenging [6, 16, 17]. Ferrimagnets provide an attractive intermediate platform that combines the advantages of both: their magnetic sublattices are coupled antiparallel, as in antiferromagnets, but unequal sublattice moments produce a finite net magnetization, allowing ferrimagnets to simultaneously host high-frequency exchange-driven collective excitations and strong magneto-optical responses [16, 18–20]. In particular, rare-earth transition-metal ferrimagnets exhibit rich spin dynamics arising from the interplay between exchange interaction, magnetic anisotropy, and spin-orbit coupling [21–28]. A defining characteristic of such systems is the magnetization compensation temperature T_{MC} , at which the opposing sublattice moments cancel, and the net magnetization vanishes. Near T_{MC} , the system acquires antiferromagnet-like properties – strongly suppressed stray fields while retaining finite magneto-optical responses, which offer a highly tunable platform for ultrafast spin control.

Among various ultrafast excitation techniques, THz radiation has emerged as a powerful and direct tool for manipulating spin systems. Coherent spin dynamics in the THz frequency range can be generated using both optical and THz excitation schemes. Optical excitation provides an indirect route to spin dynamics via mechanisms such as the inverse Faraday effect (IFE) [29, 30], in which a circularly polarized ultrafast optical pulse generates a transient effective magnetic field and initiates coherent magnetization precession. Other optical mechanisms, including the Cotton-Mouton effect [31], ultrafast heating, and subsequent modification of magnetic anisotropy or exchange interactions, can also launch coherent spin dynamics through an impulsive change in the magnetic free energy. These approaches provide femtosecond temporal resolution and have been widely employed to investigate ultrafast magnetization dynamics. However, optical excitation is generally non-resonant with respect to THz-frequency magnetic modes, and the efficiency and character

of the excitation depend on the specific light-matter coupling mechanism.

In contrast, THz radiation provides a more direct and resonant means of exciting coherent spin dynamics. The magnetic-field component of a THz pulse couples directly to magnetic moments via the Zeeman interaction and can drive collective spin modes with resonance frequencies in the THz range. Single-cycle THz pulses are particularly suitable because their broad spectral bandwidth provides good overlap with magnetic resonances while retaining sub-picosecond temporal resolution. This allows both resonant coherent excitation of exchange modes and investigation of their nonlinear response using intense THz fields [32]. THz excitation can therefore provide a direct means of accessing the intrinsic exchange-driven dynamics of antiferromagnetic and ferrimagnetic systems, complementing the indirect excitation mechanisms available through optical pumping. The temporal evolution of such dynamics is commonly described by the Landau-Lifshitz-Gilbert (LLG) equation [33–35], which captures both the precessional motion of the magnetization around an effective field and its relaxation through Gilbert damping. In rare-earth iron garnets, the THz Zeeman interaction resonantly drives a high-frequency collective spin precession between the rare-earth and iron sublattices — the Kaplan-Kittel exchange mode — which serves as the primary spectral observable throughout this thesis.

The conventional LLG formalism has been remarkably successful in describing ultra-fast spin dynamics across a wide variety of magnetic systems [36–38]. Nevertheless, in regimes of strong damping, due to high-frequency exchange oscillations, and pronounced spin-orbit coupling, it becomes insufficient to fully capture the underlying physics, and relativistic corrections to spin dynamics can become significant [39–43]. One such contribution is the field-derivative torque (FDT) – a relativistic torque arising from the time derivative of the applied electromagnetic field, whose microscopic origin lies in spin-orbit coupling and relativistic corrections to the spin Hamiltonian [20, 22, 44]. Unlike the Zeeman torque, which depends on the instantaneous value of the driving field, the FDT depends on the time

derivative of the driving field and therefore becomes particularly significant for ultrashort THz pulses. Theoretical predictions indicate that FDT effects are most pronounced in systems combining high Gilbert damping, THz-frequency magnetic resonances, and equivalent sublattice gyromagnetic ratios. Crucially, its observability is strongly material-dependent: systems with low damping, such as the antiferromagnet YFeO_3 [36] or the ferrimagnetic garnet $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ [20], fall below the threshold at which FDT effects become discernible. Despite longstanding theoretical predictions, direct experimental evidence of the relativistic FDT in any magnetic system had remained elusive prior to this work.

Motivated by these considerations, this thesis focuses on the ultrafast THz magnetization dynamics of the rare-earth iron garnet $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ (GdYb-BIG) – a ferrimagnetic system in which the net rare-earth magnetization couples antiferromagnetically to the iron magnetization, giving rise to the Kaplan-Kittel exchange mode at 0.48 THz at room temperature [45–47]. GdYb-BIG additionally exhibits a comparatively large Gilbert damping parameter and equivalent sublattice gyromagnetic ratios, making it uniquely suitable for investigating relativistic contributions to spin dynamics. Using THz-pump optical-probe spectroscopy and nonlinear 2D THz spectroscopy, we demonstrate that the observed $\chi^{(3)}$ nonlinear magnetic response cannot be reproduced by the conventional Zeeman torque alone, and that incorporating FDT into the LLG framework is essential to describe the experimental observations [48, 49]. Furthermore, the strong spin-orbit coupling of the Yb^{3+} ions introduces low-energy crystal electric field (CEF) transitions resonant with THz frequencies [50, 51], and the material hosts a magnetization compensation temperature at 96 K. Temperature-dependent THz spectroscopy reveals an unexpected hybridization between localized Yb-ion CEF excitations and the collective exchange dynamics near T_{MC} , accompanied by an anomalous redshift of the exchange resonance upon cooling – a striking reversal of the conventional blue-shift driven by strengthening exchange interactions at low temperatures, which we attribute to a CEF-mediated renormalization of the Yb-Fe exchange

anisotropy.

The present thesis is structured as follows:

Chapter 2 introduces the scientific background necessary to appreciate the results presented in the thesis. It covers the principal classes of magnetic order and the microscopic interactions that stabilize them; the Landau-Lifshitz-Gilbert equation and its relativistic extension incorporating the field-derivative torque; and the quantum-mechanical magnon picture of collective spin excitations.

Chapter 3 presents the experimental methods employed throughout this thesis: inverse Faraday effect spectroscopy, THz-pump NIR-probe spectroscopy in the Faraday geometry, linear THz time-domain spectroscopy, and nonlinear two-dimensional THz spectroscopy.

Chapter 4 is devoted to the experimental realization of the relativistic field-derivative torque in GdYb-BIG. Using THz-pump optical-probe and 2D THz spectroscopy, we demonstrate that the observed $\chi^{(3)}$ nonlinear magnetic response, comprising pump-probe and echo signals, cannot be reproduced by conventional Zeeman torque alone, and that incorporating FDT into the LLG framework is essential to describe the experimental observations.

Chapter 5 develops a theoretical understanding of the material-dependent aspects of FDT-driven nonlinear magnetization dynamics. Through systematic numerical modeling based on coupled FDT-modified LLG equations, we identify how parameters such as the sublattice spin-volume ratio and Gilbert damping govern the emergence and observability of the $\chi^{(3)}$ nonlinear THz signals, establishing design criteria for future materials in which relativistic spin-torque effects may be accessed.

Chapter 6 explores the temperature-dependent exchange dynamics and CEF-mediated coupling in GdYb-BIG across its magnetic compensation temperature. Using temperature-dependent THz time-domain spectroscopy, we demonstrate strong hybridization between the Yb-ion CEF excitations and the collective Yb-Fe exchange mode, observe a pronounced

redistribution of spectral weight near T_{MC} , and report an anomalous redshift of the exchange resonance upon cooling, which we trace to a CEF-mediated renormalization of the Yb-Fe exchange anisotropy.

Chapter 7 summarizes the findings and provides a broader outlook on the results obtained in this thesis.

Chapter 2

Scientific background

The manipulation of magnetic order at ultrafast timescales requires a thorough understanding of how spins are arranged in their equilibrium ground state and how they respond dynamically to time-dependent external perturbations. This chapter lays the theoretical framework underlying all the experimental and numerical investigations presented in this thesis. We begin by reviewing various forms of magnetic ordering and the underlying microscopic interactions for stabilizing them. We then discuss the classical LLG equation, which governs the precessional dynamics and damping of magnetic moments in an effective magnetic field. The discussion is subsequently extended to relativistic corrections of the LLG framework, with particular focus on field-derivative torque. Finally, we conclude with a quantum-mechanical perspective on collective spin excitations, introducing the magnon picture.

2.1 Magnetic ordering

The physical properties of magnetic materials are determined by the arrangement of the magnetic moments in their ground state. Depending on the nature and strength of the interactions between neighboring spins, different forms of magnetic ordering can emerge. Among them, ferromagnetic and antiferromagnetic orderings constitute the two primary classes of magnetically ordered systems. The competition among exchange interactions, thermal fluctuations, and magnetic anisotropy gives rise to a wide variety of magnetic phases that ultimately dictate the material's dynamical behavior. Understanding the different types of magnetic ordering is therefore essential before discussing the microscopic

interactions and the corresponding spin dynamics.

2.1.1 Ferromagnet

Ferromagnetism is the most familiar form of magnetic ordering and is characterized by the parallel alignment of neighboring spins below a critical temperature known as the Curie temperature (T_C); see Figure 2.1. This parallel arrangement arises from exchange interaction, which lowers the system's total energy when the spins are aligned in the same direction. As a consequence, a spontaneous magnetization exists even in the absence of an external magnetic field.

The exchange interaction responsible for ferromagnetism can be described phenomenologically by the Weiss molecular field theory. According to this model, each magnetic moment experiences an effective field generated by its neighboring spins, known as the molecular field. Below the Curie temperature, the molecular field overcomes thermal fluctuations and stabilizes a state with long-range magnetic order. The total magnetization of a ferromagnet can be expressed as

$$\mathbf{M} = \sum_i \mathbf{m}_i, \quad (2.1)$$

where $\mathbf{m}_i$ denotes the magnetic moment associated with the i^{th} atom. Since the magnetic moments are aligned parallel to each other, the net magnetization remains finite [52].

From the viewpoint of spin dynamics, ferromagnets are advantageous because finite magnetization enables efficient excitation and detection of magnetic precession using various experimental techniques, such as ferromagnetic resonance spectroscopy (FMR) and time-resolved Faraday and Kerr spectroscopy. However, the resonance frequencies in ferromagnetic systems are typically limited to the GHz regime because the restoring force is primarily determined by relatively weak anisotropy fields. Consequently, the operation speed of conventional ferromagnetic devices is restricted, motivating the search for systems possessing faster spin dynamics [53, 54].

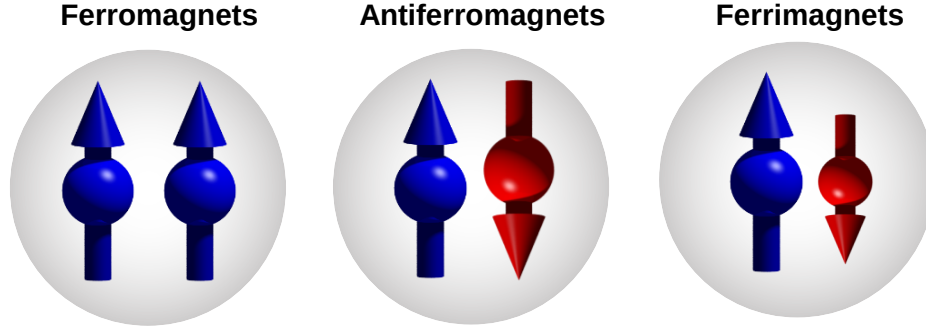


Figure 2.1: Schematic illustration of different magnetic orders: ferromagnetic, antiferromagnetic, and ferrimagnetic. The blue and red arrows denote the sublattice-dependent magnetic moment.

2.1.2 Antiferromagnet

In antiferromagnetic materials, neighboring magnetic moments align antiparallel due to a negative exchange interaction; see Figure 2.1. As a result, the total magnetization vanishes in equilibrium despite the presence of long-range magnetic order. Antiferromagnetism was first introduced by Néel, who showed that the magnetic lattice can be divided into two interpenetrating sublattices with opposite magnetizations.

For a two-sublattice antiferromagnet, the net magnetization is given by $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, where $|\mathbf{M}_1| = |\mathbf{M}_2|$, and the two sublattice magnetizations are oriented antiparallel to each other, resulting $\mathbf{M} = 0$. Although the net magnetization vanishes, the magnetic order can be described by the Néel vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$, which acts as the order parameter in the antiferromagnetic phase. Unlike ferromagnets, the dominant restoring force in antiferromagnets originates from the strong exchange interaction between the sublattices. Consequently, antiferromagnets exhibit resonance frequencies in the THz regime, making them promising candidates for ultrafast spintronic applications.

Despite the advantages, the absence of a net magnetization makes the excitation and detection of antiferromagnetic dynamics experimentally challenging [3, 20, 55]. These difficulties have motivated extensive investigations of ferrimagnetic systems, which combine

several desirable properties of both ferromagnets and antiferromagnets.

2.1.3 Ferrimagnet

When the antiferromagnetically coupled sublattices are nonequivalent, their magnetizations differ in magnitude and, therefore, do not cancel completely. The resulting magnetic state, known as ferrimagnetism (see Figure 2.1), possesses a finite net magnetization while retaining the strong exchange interaction characteristic of antiferromagnets. This combination makes ferrimagnets an ideal platform for studying ultrafast magnetization dynamics. For a two-sublattice ferrimagnet, the net magnetization is given by $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, with $|\mathbf{M}_1| \neq |\mathbf{M}_2|$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ remain oriented antiparallel to each other, but their unequal magnitudes yield $\mathbf{M} \neq 0$. As in the antiferromagnetic case, the antiparallel order can additionally be described by the Néel vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$, which remains a meaningful order parameter alongside the net magnetization $\mathbf{M}$. Because of the molecular field, the temperature dependence of the spontaneous magnetization differs between the two sublattices. Consequently, the net magnetization of a ferrimagnet can exhibit a markedly non-trivial thermal evolution [10, 18, 29, 30, 56]. In contrast to ferromagnets, whose susceptibility follows the Curie-Weiss law, the magnetic susceptibility of a ferrimagnet deviates from this because of the sublattice asymmetry. In certain materials, for example, rare-earth garnets, one sublattice magnetization dominates at low temperature while the other dominates at high temperature; since the two contributions have opposite signs, the net magnetization passes through zero at an intermediate temperature known as the magnetization compensation temperature, T_{MC} [57, 58]. In addition to this, there is another characteristic temperature where the angular momentum of the sublattices compensates, i.e., $\sum_i \frac{M_i}{g_i} = 0$, where g_i is the sub-lattice gyromagnetic ratio. This temperature point is known as the angular momentum compensation point T_{AC} [59, 60]. In the vicinity of T_{MC} , the ferrimagnet effectively mimics an antiferromagnet, with strongly suppressed net magnetization and stray field, while

retaining the underlying exchange-driven sublattice dynamics [50, 61–65]. This dual character is what makes ferrimagnets uniquely attractive for ultrafast spin dynamics [20, 66].

In these systems the strong exchange interaction between the sublattices gives rise to collective modes in which the two sublattice magnetizations can precess either predominantly in phase or out of phase. The in-phase motion corresponds to a ferromagnetic-like mode and is governed mainly by the external magnetic field, magnetic anisotropy, and demagnetization fields. In contrast, the out-of-phase motion involves a strong relative motion of the two sublattices and is governed predominantly by the inter-sublattice exchange interaction. This exchange-dominated mode is therefore often referred to as an exchange mode or antiferromagnetic-like mode. In ferrimagnetic systems, the unequal sublattice magnetizations provide an additional degree of control over the exchange dynamics. In particular, the relative magnitudes of the sublattice magnetizations and their gyromagnetic ratios influence the exchange-mode frequency, mode character, and coupling to an external magnetic field. Near a magnetization compensation point, where the two sublattice magnetizations approach equal magnitude, the character of the collective dynamics can change substantially. These properties make antiferromagnetically coupled ferrimagnets an important platform for studying high-frequency and nonlinear magnetization dynamics.

2.2 Magnetic interactions

The spin ordering described above emerges from the interplay of several distinct microscopic interactions. Understanding each of these interactions is essential for constructing the free energy density that enters the LLG equation, which governs the magnetization dynamics in all magnetically ordered systems.

2.2.1 Magnetic dipolar interaction

A natural starting point for considering the interactions between magnetic moments is the classical magnetic dipolar interaction. For two magnetic moments $\boldsymbol{\mu}_1$ and $\boldsymbol{\mu}_2$ separated by a distance $\mathbf{r}$, the dipolar interaction energy is given by

$$E_{\text{dip}} = \frac{\mu_0}{4\pi r^3} \left[\boldsymbol{\mu}_1 \cdot \boldsymbol{\mu}_2 - \frac{3}{r^2} (\boldsymbol{\mu}_1 \cdot \mathbf{r})(\boldsymbol{\mu}_2 \cdot \mathbf{r}) \right] \quad (2.2)$$

where μ_0 is the permeability of free space. This interaction is long-ranged and is strongly anisotropic, since its sign and magnitude depend on the orientation of the two moments relative to $\hat{\mathbf{r}}$. A simple order-of-magnitude estimate, obtained by setting $\mu_1 = \mu_2 = \mu_B$ (the Bohr magneton) and r equal to a typical interatomic spacing of a few Å, shows that the dipolar energy corresponds to a temperature scale of only about 1 K. This is far too small to account for the magnetic ordering observed in most ferromagnets and antiferromagnets, whose ordering temperatures can reach hundreds or even thousands of Kelvin [52]. The magnetic dipolar interaction is therefore generally too weak to be the origin of long-range magnetic order in solids, and a different, fundamentally quantum mechanical interaction - the exchange interaction must be invoked, as discussed in the following section.

2.2.2 Heisenberg exchange interaction

The exchange interaction is the most fundamental interaction responsible for magnetic ordering in solids. Unlike the dipolar interaction, which has a classical origin, the exchange interaction is purely quantum mechanical and arises from the indistinguishability of electrons together with the Coulomb interaction between them. The microscopic origin of exchange can be understood by considering two neighboring electrons occupying atomic orbitals ψ_a and ψ_b . Since electrons are fermions, the total two-electron wavefunction must be antisymmetric under the exchange of the two particles, as required by the Pauli exclusion principle.

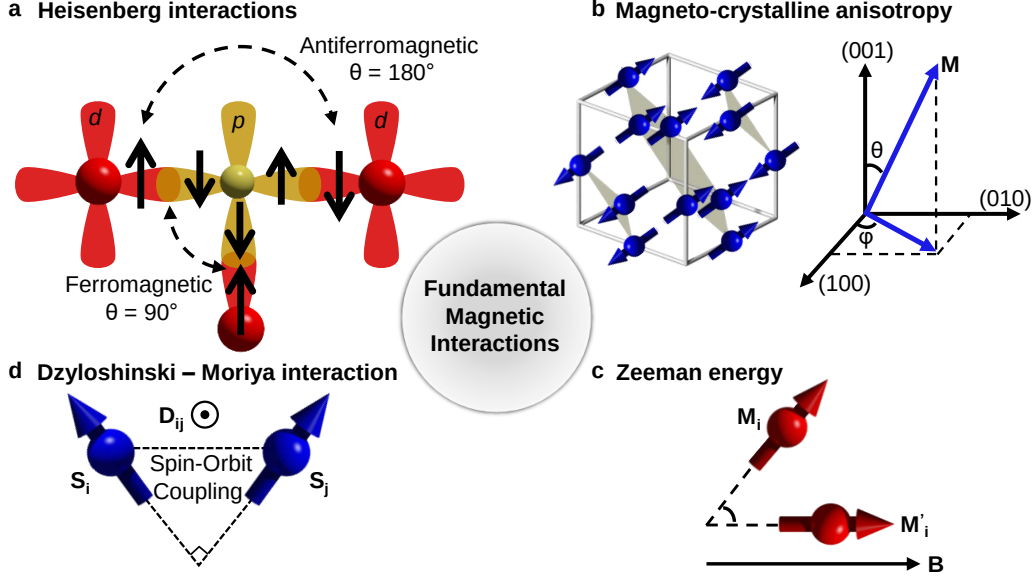


Figure 2.2: Schematic representation of the four fundamental magnetic interactions, including (a) Heisenberg exchange interaction, (b) Magneto-crystalline anisotropy, (c) Zeeman interaction with external magnetic field ($\mathbf{B}$), and (d) Dzyaloshinskii - Moriya interaction.

Depending on the symmetry of the spin state, the spatial part of the wavefunction assumes different forms. For a singlet spin state ($S = 0$), the spin wavefunction (χ_S) is antisymmetric and therefore the spatial wavefunction must be symmetric. Conversely, for a triplet state ($S = 1$), the spin wavefunction is symmetric (χ_T), requiring an antisymmetric spatial wavefunction. The corresponding total wavefunctions can be written as

$$\Psi_S = \frac{1}{\sqrt{2}} [\psi_a(\mathbf{r}_1)\psi_b(\mathbf{r}_2) + \psi_a(\mathbf{r}_2)\psi_b(\mathbf{r}_1)] \chi_S, \quad (2.3)$$

$$\Psi_T = \frac{1}{\sqrt{2}} [\psi_a(\mathbf{r}_1)\psi_b(\mathbf{r}_2) - \psi_a(\mathbf{r}_2)\psi_b(\mathbf{r}_1)] \chi_T. \quad (2.4)$$

The energies ($E_{S,T}$) corresponding to these two configurations are obtained from the expectation value of the electronic Hamiltonian,

$$E_{S,T} = \int \Psi_{S,T}^* \hat{\mathcal{H}} \Psi_{S,T} d\mathbf{r}_1 d\mathbf{r}_2. \quad (2.5)$$

The difference between these singlet and triplet energies arises from the exchange symmetry of the electronic wavefunctions. This energy difference can be expressed in terms of the exchange integral,

$$J = \frac{E_S - E_T}{2}, \quad (2.6)$$

which quantifies the strength of the exchange interaction between neighboring spins. The sign of J determines the preferred magnetic configuration. When $J > 0$, the triplet state possesses lower energy, favoring parallel alignment of neighboring spins and resulting in ferromagnetic ordering. On the other hand, when $J < 0$, the singlet state becomes energetically favorable, leading to antiparallel spin alignment and antiferromagnetic ordering.

Using the exchange integral, the spin-dependent part of the electronic Hamiltonian can be written in the compact form

$$\hat{\mathcal{H}}_{\text{ex}} = -2J\mathbf{S}_1 \cdot \mathbf{S}_2, \quad (2.7)$$

which is known as the Heisenberg exchange Hamiltonian. For a magnetic crystal containing a large number of interacting spins, the Hamiltonian is generalized as

$$\hat{\mathcal{H}}_{\text{ex}} = - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2.8)$$

where the summation extends over neighboring spin pairs and J_{ij} denotes the exchange constant between the i^{th} and j^{th} spins [52].

Direct exchange interaction

The simplest realization of the exchange interaction described above is direct exchange, in which the coupling between two neighboring magnetic moments arises from the overlap of their electronic wavefunctions without the mediation of any intervening atom. This is

the most straightforward way in which the anti-symmetrization argument of the previous section translates into an effective spin-spin coupling: when the unpaired electrons on two adjacent ions are close enough for their orbitals to overlap appreciably, the Coulomb repulsion between them, combined with the Pauli exclusion principle, directly sets the energy difference between the parallel and antiparallel spin configurations. Direct exchange is consequently very short-ranged and decays rapidly with interatomic separation. In particular, this limits the applicability of direct exchange in most magnetic insulators, including the rare-earth iron garnets considered in this thesis, where the magnetic ions occupying different sublattices are typically too far apart for their orbitals to overlap directly [52].

Indirect exchange interaction

Thus, in many magnetic insulators, the exchange interaction is mediated indirectly, typically through a non-magnetic intermediary. The most important indirect exchange mechanism in magnetic oxides is superexchange, in which two magnetic cations interact via a non-magnetic anion - commonly oxygen situated between them; see Figure 2.2a. Although the anion itself carries no permanent magnetic moment, virtual hopping of an electron from the anion's filled p orbital into the empty or partially filled d orbital of a neighboring magnetic cation, and back again, allows the spin configuration on one cation to influence the spin configuration on the other. This virtual transfer process lowers the total energy of the system, thereby setting in an effective exchange coupling between them despite the absence of any direct orbital overlap. The sign and strength of the superexchange interaction depend strongly on the cation-anion-cation bond geometry. When the bond angle is close to 180° , the same anion p orbital overlaps with the relevant d orbitals of both magnetic cations along a common axis, allowing virtual electron transfer between two half-filled cation orbitals. The Pauli exclusion principle then forbids this transfer unless the two cation spins are antiparallel, so that 180° superexchange paths give rise to strong antiferromagnetic coupling;

see Figure 2.2a. By contrast, when the bond angle is close to 90° , the two cation orbitals overlap with two mutually orthogonal anion p orbitals rather than a shared one, and the virtual transfer instead favors parallel alignment of the cation spins via an intra-atomic exchange process on the anion, resulting in a comparatively weaker ferromagnetic coupling. This geometric dependence makes superexchange a particularly versatile mechanism for setting in the ground state of oxide materials [52, 67]. In the rare-earth iron garnets studied in this thesis, the antiferromagnetic coupling between the iron and rare-earth sublattices is mediated by such oxygen-bridged superexchange paths that ultimately stabilize the ferromagnetic ground state of GdYb-BIG.

2.2.3 Magneto-crystalline anisotropy energy

While the Heisenberg exchange interaction determines whether neighboring spins prefer to align parallel or antiparallel, it is isotropic by nature: the scalar product $\mathbf{S}_i \cdot \mathbf{S}_j$ depends only on the relative angle between the two spins and not on their orientation with respect to the crystal lattice. In real magnetic materials, however, the magnetization is rarely free to point in an arbitrary direction; instead, it is energetically favorable for it to align along certain preferred crystallographic directions, known as easy axes. This directional dependence of the magnetic energy is referred to as magnetic anisotropy, and its microscopic origin lies in spin-orbit coupling, which links the electron spin to its orbital motion [67, 68]. Because spin-orbit coupling is a relativistic effect, the resulting anisotropy energy is typically one to two orders of magnitude smaller than the exchange energy, yet it has a profound influence on the static and dynamic properties of a magnetic material. The simplest and most common form of magnetic anisotropy relevant to the rare-earth iron garnets studied in this thesis is uniaxial anisotropy, see Figure 2.2b, in which the energy depends on the orientation of the magnetization $\mathbf{M}$ relative to a single preferred axis $\hat{\mathbf{n}}$. The corresponding anisotropy energy

density can be written as

$$\mathcal{F}_{\text{ani}} = -K \frac{(\mathbf{M} \cdot \hat{\mathbf{n}})^2}{|\mathbf{M}|^2} \quad (2.9)$$

where K is the uniaxial anisotropy constant. For $K > 0$, the energy is minimized when $\mathbf{M}$ is parallel or antiparallel to $\hat{\mathbf{n}}$, defining an easy axis of magnetization. For $K < 0$, the energy is instead minimized when $\mathbf{M}$ lies anywhere within the plane perpendicular to $\hat{\mathbf{n}}$, defining an easy plane. The quadratic dependence on $(\mathbf{M} \cdot \hat{\mathbf{n}})$ reflects the fact that anisotropy energy must remain invariant under time reversal, $\mathbf{M} \rightarrow -\mathbf{M}$, ruling out odd terms in the magnetization. For example, Figure. 2.2b illustrates the crystal unit cell containing a single magnetic moment, with the easy axis magnetization oriented along the $[111]$ crystallographic direction. In GdYb-BIG, for instance, the iron and rare-earth sublattices are each assigned a distinct uniaxial anisotropy constant, K_{Fe} and K_{RE} respectively, with the shared easy axis oriented along the $[111]$ crystallographic direction.

2.2.4 Zeeman energy

When a magnetic moment is placed in an external magnetic field, it experiences an interaction energy that depends on the relative orientation between the moment and the field - this is known as the Zeeman interaction. Physically, the field exerts a torque on the magnetic moment, favoring an alignment in which the moment points along the field direction, since this configuration minimizes the system's energy. The energy density for the Zeeman term can be expressed as

$$\mathcal{F}_Z = -\mu_0 \mathbf{M} \cdot \mathbf{B} \quad (2.10)$$

$\mathbf{B}$ is the applied magnetic field, see Figure 2.2c. Unlike the exchange interaction, which is intrinsic to the material and fixed by its electronic and crystal structure, the Zeeman energy depends entirely on an externally applied field and therefore vanishes in its absence. It is not a property of the material itself, but rather the principal means by which an external agent,

whether a static magnet or a time-varying electromagnetic field, can be used to manipulate, drive, or probe an otherwise equilibrium magnetic system. While in the presence of a static field, the Zeeman term competes with the exchange and anisotropy energies to select a particular equilibrium orientation of the magnetization, in the presence of a time-dependent field, it instead exerts a continuously varying torque that drives the magnetization out of equilibrium and induces precessional motion. This is the starting point for nearly all forms of field-driven magnetization dynamics.

2.2.5 Demagnetization energy

In addition to the exchange, crystalline anisotropy, and Zeeman energies, the magnetization of a ferromagnetic or ferrimagnetic body is also influenced by the demagnetization energy, which originates from the magnetic field produced by the material itself. When a system is magnetized, magnetic poles appear at its surfaces, generating an internal magnetic field, known as the demagnetizing field ($\mathbf{H}_d$), that opposes the magnetization. The associated energy can be expressed as

$$E_{\text{demag}} = -\frac{1}{2}\mu_0 \int_V \mathbf{M} \cdot \mathbf{H}_d, dV, \quad (2.11)$$

where μ_0 is the permeability of free space, $\mathbf{M}$ is the magnetization, and the integration is performed over the sample volume. Since the demagnetizing field depends strongly on the sample geometry, the demagnetization energy is often referred to as the shape anisotropy energy. Magnetic systems naturally tend to minimize this energy by forming magnetic domains or by aligning the magnetization along directions for which the demagnetizing field is smallest. Consequently, the demagnetization energy plays an important role in determining the equilibrium magnetic configuration and strongly influences the magnetization dynamics [52].

For bulk ferrimagnetic iron garnets, the demagnetizing field can be comparable in mag-

nitude to the externally applied magnetic field and therefore cannot be neglected while describing the spin dynamics. In the GdYb-BIG system investigated in this thesis, the demagnetizing field is approximately 110 mT, which is nearly equal to the applied static magnetic field of 120 mT used in the experiments. As a result, the effective magnetic field experienced by the magnetic sublattices is determined by the combined contributions of the external field, exchange interaction, magnetic anisotropy, and the demagnetizing field.

2.2.6 Dzyaloshinskii-Moriya interaction

All the interactions discussed so far are symmetric under the exchange of the two coupled spins. This means the energy depends only on the relative angle between $\mathbf{S}_i$ and $\mathbf{S}_j$ and is invariant under $\mathbf{S}_i \leftrightarrow \mathbf{S}_j$. In certain magnetic crystals, however, an additional, fundamentally different exchange term can arise: the Dzyaloshinskii-Moriya interaction (DMI), an *antisymmetric* exchange coupling of the form

$$\mathcal{H}_{\text{DMI}} = -\mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (2.12)$$

where $\mathbf{D}_{ij}$ is the Dzyaloshinskii-Moriya vector. Unlike the Heisenberg term, the cross product in Eq. (2.12) changes sign under $\mathbf{S}_i \leftrightarrow \mathbf{S}_j$, so this interaction does not simply favor parallel or antiparallel spin alignment; instead, it favors a perpendicular, canted arrangement of the two spins, with the sense of canting (clockwise or counterclockwise) fixed by the direction of $\mathbf{D}_{ij}$, see Figure 2.2d. This interaction was first proposed by Dzyaloshinskii on symmetry grounds, as a means of explaining the small canted moment observed in certain antiferromagnetic crystals, and was subsequently given a microscopic derivation by Moriya, who showed that such an antisymmetric term arises naturally as a relativistic correction to Anderson's superexchange theory [67]. The microscopic origin of the DMI is rooted in the combined presence of spin-orbit coupling and broken spatial inversion symmetry at the relevant magnetic site [52, 67, 68]. Although the DMI is not the dominant

interaction governing the collinear ferrimagnetic ground state of GdYb-BIG studied in this thesis, it is included here for completeness.

2.3 Magnetization dynamics

Having established the principal interactions that determine the equilibrium magnetic structure of a material, we now turn to its dynamical response: how the magnetization evolves in time once it is displaced from equilibrium by an external perturbation. The starting point for this discussion is the torque on a magnetic moment in an effective magnetic field, which gives rise to precessional motion.

2.3.1 Landau-Lifshitz-Gilbert equation

The temporal evolution of magnetization in a magnetic material is conventionally described using phenomenological equations of motion. The earliest and most widely used description was proposed by Landau and Lifshitz, who considered the dynamics of a magnetic moment under the influence of an effective magnetic field. Within this framework, the magnetization vector undergoes precessional motion about the effective field while simultaneously undergoing transverse relaxation that drives the system towards its equilibrium state. Since the damping torque acts perpendicular to the magnetization vector, the magnitude of the magnetization remains conserved during the dynamics. For a magnetization vector ($\mathbf{M}(\mathbf{r}, t)$), the Landau-Lifshitz (LL) equation can be written as [63]

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{B}^{\text{eff}} - \frac{\lambda}{|\mathbf{M}|} \mathbf{M} \times (\mathbf{M} \times \mathbf{B}^{\text{eff}}), \quad (2.13)$$

where γ denotes the gyromagnetic ratio, λ is the phenomenological damping constant, and $\mathbf{B}^{\text{eff}}$ represents the effective magnetic field acting on the magnetization. The effective field generally contains contributions from the externally applied magnetic field, exchange interactions, magnetic anisotropy, dipolar interactions, and demagnetizing fields [69–71].

Although the LL equation successfully describes many aspects of magnetization dynamics, it becomes inadequate for materials exhibiting relatively large damping. To overcome this limitation, Gilbert introduced an alternative formulation in which the dissipative torque is proportional to the time derivative of the magnetization itself. The resulting Landau-Lifshitz-Gilbert equation is expressed as [35]

$$\frac{d\mathbf{M}}{dt} = -\gamma' \mathbf{M} \times \mathbf{B}^{\text{eff}} + \alpha \mathbf{M} \times \frac{d\mathbf{M}}{dt}, \quad (2.14)$$

Despite their different mathematical forms, LL and LLG equations are mathematically equivalent and can be transformed into one another through appropriate redefinitions of the gyromagnetic ratio and damping parameter. The corresponding relations are given by $\gamma = \frac{\gamma'}{1+\alpha^2}$, and $\lambda = \frac{\alpha\gamma'}{1+\alpha^2}$ where γ' is the gyromagnetic ratio appearing in the Gilbert formulation, γ is the effective gyromagnetic ratio in the LL equation, and α is the dimensionless Gilbert damping parameter.

The dynamics described by these equations therefore consist of two fundamental processes: precession of the magnetization about the effective field and damping that gradually aligns the magnetization with the equilibrium direction; see Figure 2.3. Originally, the damping parameter was regarded as a scalar quantity and was generally attributed to relativistic effects arising from spin-orbit coupling [21–28, 72].

Over the years, the LLG framework has been extended further to account for additional dynamical phenomena that become important on ultrafast timescales. For example, the inclusion of a second-order time derivative of the magnetization gives rise to magnetic inertia or spin nutation [43, 73–75], whereas incorporating spin-current terms allows one to describe current-induced torques such as spin-transfer torque [76–79] and spin-orbit torque [55, 80–84]. These extensions highlight that the conventional LLG equation, although remarkably successful, represents only the lowest-order description of magnetiza-

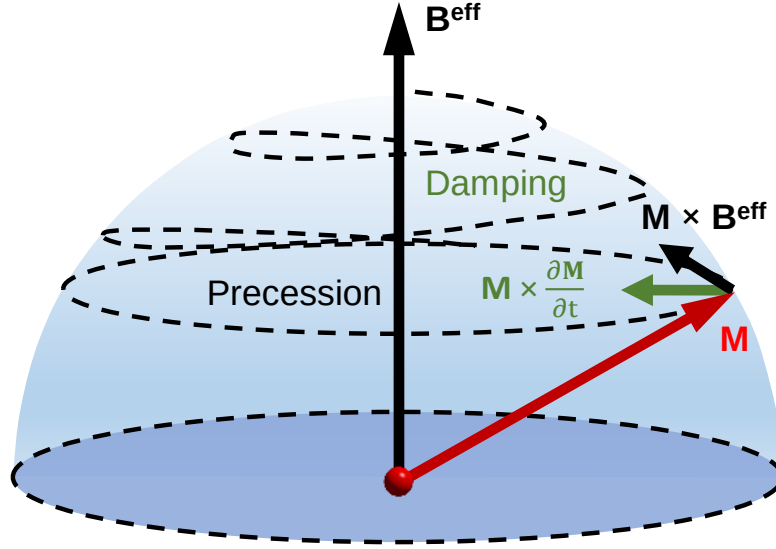


Figure 2.3: Schematic depiction of Magnetization dynamics. The red arrow shows the magnetization vector. It precesses around the effective field, and the damping curves are shown in the black dashed line.

tion dynamics and requires further modifications when relativistic and nonequilibrium effects become significant.

2.3.2 Relativistic field-derivative torque

The LLG equation has been remarkably successful in describing magnetization dynamics in a wide variety of magnetic systems. However, when the magnetic system is driven by ultrashort electromagnetic pulses, particularly in the THz frequency regime, the conventional LLG formalism may no longer provide a complete description of the spin dynamics. This is because the derivation of the Gilbert damping assumes that the magnetization responds to a harmonic excitation with a well-defined frequency. In contrast, a single-cycle THz pulse is inherently non-harmonic and contains a broad range of frequency components. Under such conditions, additional relativistic contributions to the magnetization dynamics can become important.

The origin of the field-derivative torque can be traced back to the relativistic spin-orbit

interaction between the electron spin and the external electromagnetic field [22]. The external spin-orbit Hamiltonian can be written as

$$H_{\text{SOC}}^{\text{ext}} = -\frac{e}{4m^2c^2} \mathbf{S} \cdot [\mathbf{E}_{\text{ext}} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathbf{E}_{\text{ext}}], \quad (2.15)$$

where $\mathbf{S}$ is the spin angular momentum, $\mathbf{p}$ is the momentum operator, $\mathbf{A}$ is the vector potential, and $\mathbf{E}_{\text{ext}}$ is the external electric field associated with the applied electromagnetic radiation. The contribution relevant to the field-derivative torque can be identified using the operator identity

$$\mathbf{p} \times \mathbf{E}_{\text{ext}} = -\mathbf{E}_{\text{ext}} \times \mathbf{p} - i\hbar \nabla \times \mathbf{E}_{\text{ext}}, \quad (2.16)$$

together with Faraday's law,

$$\nabla \times \mathbf{E}_{\text{ext}} = -\frac{\partial \mathbf{B}}{\partial t}. \quad (2.17)$$

Substitution of Eq. (2.17) into Eq. (2.16) gives

$$\mathbf{p} \times \mathbf{E}_{\text{ext}} = -\mathbf{E}_{\text{ext}} \times \mathbf{p} + i\hbar \frac{\partial \mathbf{B}}{\partial t}. \quad (2.18)$$

Consequently, the spin-orbit Hamiltonian contains a term proportional to

$$H_{\text{FDT}} \propto \mathbf{S} \cdot \frac{\partial \mathbf{B}}{\partial t}. \quad (2.19)$$

therefore giving an additional torque on the magnetization that is proportional to

$$\boldsymbol{\tau}_{\text{FDT}} \propto \mathbf{M} \times \frac{\partial \mathbf{B}}{\partial t}. \quad (2.20)$$

Thus, unlike the conventional Zeeman torque, which depends on the instantaneous magnetic field, the field-derivative torque is determined by the temporal variation of the applied magnetic field. This contribution becomes particularly relevant for ultrashort THz pulses, which contain rapidly varying electric and magnetic fields.

More generally, the relativistic treatment of magnetization dynamics leads to the following equation for a time-dependent magnetic field [22]:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} + \mathbf{M} \times \left[\bar{\mathbf{A}} \cdot \left(\frac{\partial \mathbf{M}}{\partial t} + \frac{\partial \mathbf{H}}{\partial t} \right) \right], \quad (2.21)$$

where $\bar{\mathbf{A}}$ is the relativistic damping tensor. The first term describes the conventional precessional dynamics, while the second term contains the relativistic damping and the additional contribution associated with the time dependence of the external magnetic field. For isotropic damping, the damping tensor can be reduced to a scalar Gilbert damping parameter α , and the field-dependent contribution takes the form

$$\boldsymbol{\tau}_{\text{FDT}} \propto \alpha \mathbf{M} \times \frac{\partial \mathbf{H}}{\partial t}. \quad (2.22)$$

This shows that the field-derivative torque and the conventional Gilbert damping has a common relativistic origin in the spin-orbit interaction. For the atomistic description used in this thesis, the magnetization of the i th sublattice can be expressed in terms of the dimensionless magnetization vector $\mathbf{m}_i$ as

$$\mathbf{M}_i = \frac{\mu_{s,i}}{a_i^3} \mathbf{m}_i, \quad (2.23)$$

where a_i^3 is the volume per spin of the i th sublattice and $\mu_{s,i}$ is the magnetic moment per spin. Therefore,

$$\frac{\partial \mathbf{M}_i}{\partial t} = \frac{\mu_{s,i}}{a_i^3} \frac{\partial \mathbf{m}_i}{\partial t}. \quad (2.24)$$

Using this relation in the isotropic form of the magnetization dynamics, the equation of motion for the dimensionless magnetization can be written as

$$\frac{\partial \mathbf{m}_i}{\partial t} = -\gamma_i \mathbf{m}_i \times \mathbf{B}_{\text{eff},i} + \mathbf{m}_i \times \left[\alpha_i \frac{\partial \mathbf{m}_i}{\partial t} + \frac{\alpha_i a_i^3}{\mu_{s,i}} \frac{\partial \mathbf{H}_{\text{THz}}}{\partial t} \right]. \quad (2.25)$$

Since the magnetic induction and magnetic field are related by

$$\mathbf{B}_{\text{THz}} = \mu_0 \mathbf{H}_{\text{THz}}, \quad (2.26)$$

The field-derivative contribution can be written in terms of the THz magnetic induction as

$$\frac{\partial \mathbf{H}_{\text{THz}}}{\partial t} = \frac{1}{\mu_0} \frac{\partial \mathbf{B}_{\text{THz}}}{\partial t}. \quad (2.27)$$

The FDT can therefore be incorporated into the effective magnetic field appearing in the conventional LLG equation as

$$\mathbf{B}_{\text{eff},i} \rightarrow \mathbf{B}_{\text{eff},i} - \frac{\alpha_i a_i^3}{\gamma_i \mu_{s,i} \mu_0} \frac{\partial \mathbf{B}_{\text{THz}}}{\partial t}. \quad (2.28)$$

Equation (2.28) explicitly shows the origin of the renormalization factor associated with the field-derivative torque. The proportionality to the Gilbert damping parameter α_i reflects the relativistic origin of the FDT and its connection to the same spin-orbit interaction responsible for Gilbert damping. The factor $a_i^3/\mu_{s,i}$ arises from the conversion between magnetization density and the magnetic moment of an individual spin, as given by Eq. (2.23). The factor $1/\gamma_i$ converts the additional torque into an effective magnetic field within the conventional LLG formulation, while the factor $1/\mu_0$ appears when the field derivative is expressed in terms of the magnetic induction $\mathbf{B}_{\text{THz}}$ rather than $\mathbf{H}_{\text{THz}}$.

For a simplified oscillatory magnetic field

$$\mathbf{B}_{\text{THz}}(t) = \mathbf{B}_0 \cos(2\pi\nu t), \quad (2.29)$$

its time derivative is

$$\frac{\partial \mathbf{B}_{\text{THz}}}{\partial t} = -2\pi\nu \mathbf{B}_0 \sin(2\pi\nu t). \quad (2.30)$$

Thus, the magnitude of the FDT contribution increases linearly with both the Gilbert damping parameter and the excitation frequency. This dependence explains why the FDT can become relevant for high-frequency collective spin excitations driven by intense THz pulses. In the following chapters, this relativistic correction is incorporated into the coupled LLG equations to investigate the nonlinear THz dynamics of the ferrimagnetic system.

Unlike the conventional Zeeman torque, which depends on the instantaneous value of the magnetic field, the FDT depends on how rapidly the field changes in time. Since ultra-short THz pulses possess extremely steep rising and falling edges, the magnitude of the field derivative can become sufficiently large for the FDT contribution to significantly influence the spin dynamics; see Figure 2.4a.

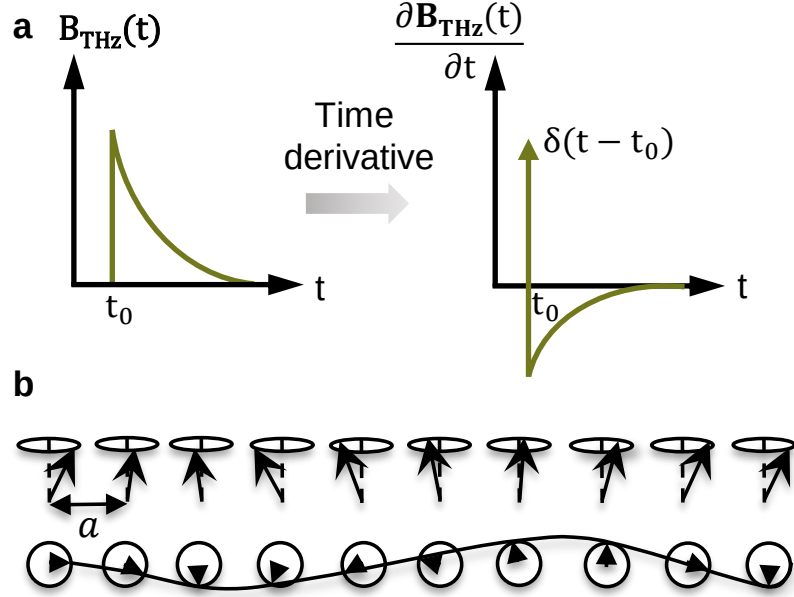


Figure 2.4: **(a)** Schematic representation of the THz magnetic field and the field derivative term. **(b)** Schematic representation of collective spin excitation in a one-dimensional crystal lattice chain. a is the lattice parameter.

Equation (2.28) shows that the magnitude of the FDT contribution depends directly on the damping parameter and the temporal variation of the applied THz field. Consequently, the effect of FDT is expected to be most pronounced in magnetic systems that simultaneously possess high damping and high-frequency collective excitations. Moreover, since differentiation introduces an additional phase into an oscillatory signal, the FDT modifies not only the amplitude of the effective driving field but also its phase relative to the conventional Zeeman field. This phase modification can have important consequences for the coherent and nonlinear magnetization dynamics.

Although the field-derivative torque was theoretically predicted several years ago, its experimental observation remained elusive because most magnetic materials studied previously have relatively small damping parameters, making the FDT contribution negligible compared to the conventional Zeeman torque. Ferrimagnetic systems, however, offer a unique opportunity in this regard. Their combination of THz-frequency exchange dy-

namics, finite net magnetization, and comparatively large Gilbert damping makes them ideal candidates for exploring relativistic corrections to the conventional LLG equation. In the following chapters, we demonstrate that the ferrimagnetic garnet $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ satisfies these requirements and provides the first experimental realization of the field-derivative torque in ultrafast magnetization dynamics.

2.4 Magnons: The quantum picture of spin precession

The classical description of magnetization dynamics in terms of spin precession can be recast within a quantum-mechanical framework by introducing spin waves. In an ordered magnetic system, a small deviation of one spin from its equilibrium orientation propagates through the lattice via exchange interactions, giving rise to a collective excitation known as a spin wave; see Figure 2.4b. Upon quantization, these spin-wave excitations are described in terms of bosonic quasiparticles called *magnons*. Each magnon carries an energy $\hbar\omega$ and a spin angular momentum of $\hbar$, corresponding to the reversal of one unit of spin relative to the ground state. Magnons, therefore, represent the fundamental elementary excitations of magnetically ordered systems and play a central role in determining their static, thermodynamic, and dynamical properties [52, 67, 68]. In ferrimagnetic systems, the strong exchange interaction between different magnetic sublattices gives rise to high-frequency magnon modes in the THz regime, which form the basis of the ultrafast magnetization dynamics investigated throughout this thesis.

Chapter 3

Experimental methods

The theoretical framework developed in Chapter 2 established that the magnetization dynamics of antiferromagnetically coupled spins can be driven, probed, and manipulated by intense THz electromagnetic fields. For the ferrimagnetic system (GdYb-BIG), studied in this thesis, the nonlinear response carries direct signatures of relativistic spin torques that are inaccessible within the conventional linear excitation regime. Translating this theoretical picture into experimental observables requires a set of complementary measurement techniques, each sensitive to a different aspect of the magnetization dynamics and operating over a different range of excitation amplitudes and timescales. This chapter describes the four experimental methods employed throughout this thesis: the inverse Faraday effect, which provides a non-resonant, impulsive means of optically orienting spins without direct thermal heating; THz-pump near-infrared-probe spectroscopy in the Faraday geometry, which gives direct time-resolved access to the out-of-plane magnetization dynamics driven by resonant THz excitation; linear THz time-domain spectroscopy, which characterizes the frequency-domain response of the exchange resonance mode and its material absorption properties; and nonlinear two-dimensional THz spectroscopy, which is the central experimental tool of this thesis and enables the $\chi^{(3)}$ nonlinear spin dynamics to be resolved in a two-dimensional frequency map, revealing the signatures of the field-derivative torque that form the basis of the results presented in Chapters 4 and 5.

3.1 Inverse Faraday effect

The magneto-optical Faraday effect describes the rotation of the polarization plane of linearly polarized light propagating through a magnetized medium, arising from the difference in refractive indices experienced by the two circularly polarized eigenmodes of the light field. The inverse of this process – the inverse Faraday effect (IFE) – refers to the generation of an effective static magnetic field, or equivalently a static magnetization, by an intense circularly polarized optical pulse propagating through a magnetic or even a non-magnetic medium [7, 85–87]. Phenomenologically, the optically induced effective magnetic field can be expressed as

$$\delta H_i^F(0) = \chi_{ijk} [E_j(\omega)E_k^*(\omega) - E_k(\omega)E_j^*(\omega)] , \quad (3.1)$$

where χ_{ijk} is a third-rank axial tensor with nonzero components for crystals of any symmetry, and $E_j(\omega)$ and $E_k(\omega)$ are the frequency-domain components of the optical electric field. The key physical consequence of Equation (3.1) is that $\delta \mathbf{H}^F$ is proportional to the vector product $\mathbf{E} \times \mathbf{E}^*$, which is nonzero only for elliptically or circularly polarized light and changes sign when the helicity is reversed from left- to right-circular polarization – a hallmark that unambiguously distinguishes the IFE from thermal effects, which are helicity-independent. The induced magnetic field is directed along the wave vector $\mathbf{k}$ of the light and acts on the magnetization during the temporal presence of the optical pulse in the material. Crucially, the IFE does not rely on optical absorption; it is a dispersive, non-dissipative process enabled by the spin-orbit coupling of the medium, making it genuinely nonthermal in character. This is in contrast to most other optical excitation mechanisms, where energy deposition and subsequent heating of the electron, spin, and lattice subsystems are unavoidable on picosecond timescales. The strength of the optically induced field is directly related to the Verdet constant of the material and can reach values of several tenths of a Tesla for

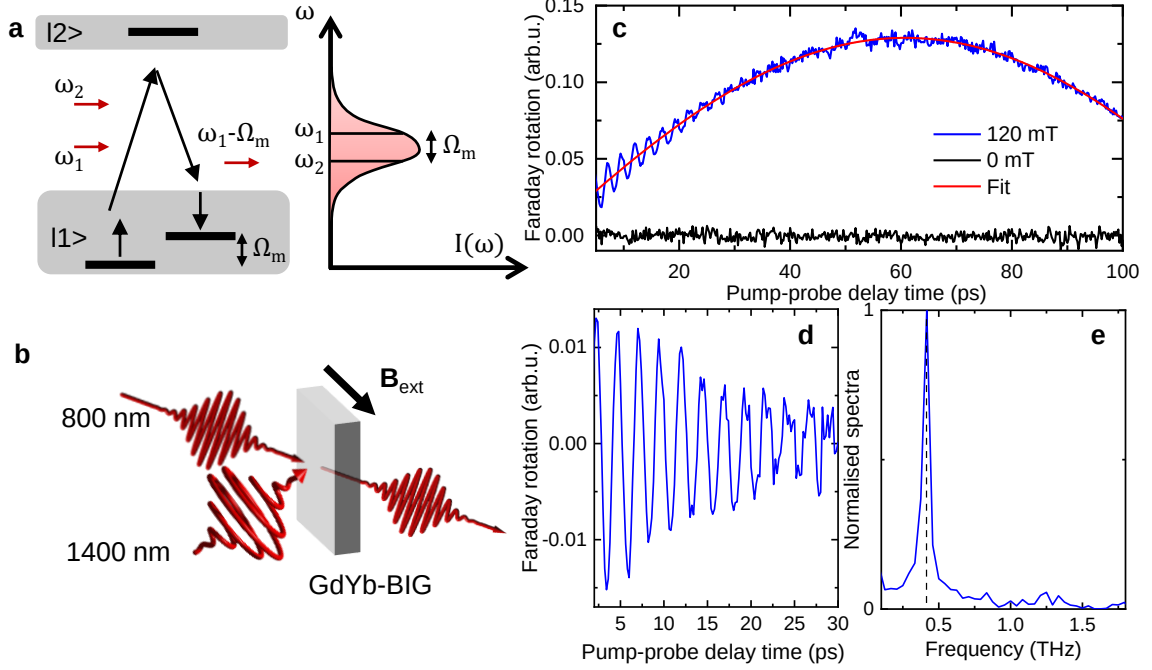


Figure 3.1: **(a)** Schematic illustration of the stimulated Raman-like coherent scattering mechanism underlying the optically induced effective magnetic field via the inverse Faraday effect. **(b)** Experimental configuration for the optical-pump optical-probe measurement in the inverse Faraday geometry. **(c)** Time-resolved Faraday rotation of the probe pulse as a function of pump-probe delay, showing a high-frequency oscillation superimposed on a slowly varying low-frequency modulation. **(d)** Extracted high-frequency oscillatory component from the transient Faraday rotation signal. **(e)** Fast Fourier transform (FFT) of the extracted oscillation, revealing the excitation of the Kaplan-Kittel exchange mode at 0.48 THz.

femtosecond laser pulses of moderate fluence in magnetic garnets.

In the microscopic picture, the IFE can be understood as a stimulated Raman-like coherent optical scattering process [87–89], in which two frequency components ω_1 and ω_2 of the spectrally broad femtosecond laser pulse participate. The component at ω_1 excites a virtual optical transition from the spin ground state to an intermediate state with strong spin-orbit coupling, while the component at ω_2 stimulates the relaxation back to the spin-split ground state with the electron spin reversed. This coherent two-photon process creates a magnon ($\hbar\Omega_m$) of energy $\hbar(\omega_1 - \omega_2)$ and is accompanied by the coherent emission of a

photon, effectively transferring angular momentum from the photon field to the spin system; see Figure 3.1a. Because the virtual state energy is of the order of the photon energy ($E = \hbar\omega \approx 1.54$ eV for 800 nm pulses), the process can be extremely fast - of the order of ≈ 3 fs - and is therefore fully consistent with the observation of magnetization changes on femtosecond timescales that cannot originate from thermal mechanisms.

In the context of this thesis, the IFE is employed as a non-resonant, impulsive excitation mechanism to investigate the spin dynamics of GdYb-BIG. A circularly polarized near-infrared pump pulse (1400 nm) generates a transient effective magnetic field through the IFE, which acts as an ultrafast perturbation to the ferrimagnetic spin system and initiates the precession of the magnetization about its equilibrium effective field, as illustrated in Figure 3.1b. Using this technique, the high-frequency Kaplan-Kittel exchange mode of GdYb-BIG is identified through time-resolved magneto-optical Faraday rotation measurements of a co-propagating probe pulse (800 nm). As shown in Figure 3.1c, the transient Faraday rotation exhibits a high-frequency oscillation superimposed on a slowly varying low-frequency modulation as a function of pump-probe delay. After isolating the high-frequency component, its Fourier transform reveals a distinct resonance at 0.48 THz, corresponding to the Kaplan-Kittel exchange mode [45–47], as shown in Figures 3.1d and e. The low-frequency modulation originates from the backward volume magnetostatic wave (BVMW) mode under an applied static magnetic field of 120 mT. Although the IFE provides an efficient non-resonant route for exciting coherent spin dynamics, it does not allow selective resonant driving of the exchange mode. This limitation motivates the use of resonant THz-pump optical-probe spectroscopy and nonlinear two-dimensional THz spectroscopy, which are discussed in the following sections.

3.2 THz-pump optical-probe spectroscopy in the Faraday geometry

3.2.1 THz radiation for spin dynamics

The optical excitation techniques discussed in the preceding section and the inverse Faraday effect in particular provide a powerful non-thermal route to driving magnetization dynamics on femtosecond timescales. However, they share a fundamental limitation: the optomagnetic field generated by a near-infrared pulse is impulsive in character, meaning it acts as a temporally brief perturbation whose spectral content is broadband and whose central frequency lies far above the intrinsic resonance frequencies of the magnetic system. This large spectral mismatch between the excitation and the spin-wave resonance results in poor spectral overlap and limits the amplitude of the coherent spin excitation that can be achieved for a given pulse fluence. Resonant excitation can overcome the limited precession amplitude of impulsive excitation by continuously driving the magnetization at its natural resonance frequency. As the spins remain phase-locked to the driving field, the precession amplitude builds up progressively over multiple precessional cycles. However, this approach compromises the temporal resolution and requires a frequency-tunable, narrowband source whose frequency matches the magnetic resonance of the material. Such a requirement is experimentally challenging, particularly for ferrimagnetic and antiferromagnetic systems, whose resonance frequencies typically lie in the THz range [32].

Intense single-cycle THz pulses offer a compelling solution that combines the advantages of both approaches while avoiding the limitations of each [32]. On the one hand, THz pulses are spectrally broad, covering a bandwidth that spans from sub-THz to several THz, and can therefore be tuned into resonance with the intrinsic spin dynamics of a wide range of magnetic materials without requiring a narrowband source. On the other hand, their temporal duration is of the order of one picosecond, comparable to or shorter than the spin-

precession period of THz-frequency exchange modes, so they retain sub-picosecond temporal resolution sufficient to resolve the full time-domain evolution of the coherent dynamics. This combination of good spectral overlap and high temporal resolution makes single-cycle THz pulses uniquely suited for investigating ultrafast spin dynamics in systems with THz-frequency resonances such as GdYb-BIG. In particular, THz radiation interacts with spins in a fundamentally more direct way than optical pulses: rather than acting through virtual electronic transitions and magneto-optical coupling effects, the magnetic field component (B_{THz}) of the THz pulse couples directly to the spin magnetic moments through the Zeeman interaction, exerting a Zeeman torque that drives the magnetization into precession in a resonant, coherent, and non-thermal manner. This direct, field-level coupling is far more efficient and controllable than the optically induced effective field of the IFE, and it allows the spin system to be driven resonantly into the nonlinear regime.

3.2.2 Experimental implementation

In the THz-pump optical-probe measurement scheme employed in this thesis, single-cycle THz pulses of peak amplitude approximately 25 mT are generated by optical rectification of 800 nm femtosecond laser pulses in a BNA-S crystal. The generated THz pulses are focused onto the GdYb-BIG crystal at normal incidence in the presence of an in-plane external static magnetic field of 120 mT, applied perpendicular to the THz magnetic field to saturate the sublattice magnetizations and establish a well-defined ferrimagnetic ground state. The THz pulse excites the magnetization dynamics of the sample via the Zeeman torque, driving the sublattice magnetizations out of their equilibrium orientations and triggering coherent precession of the Kaplan-Kittel exchange mode.

The ensuing magnetization dynamics are probed by a co-propagating, linearly polarized near-infrared (NIR) beam at 800 nm via the magneto-optical Faraday effect. The Faraday rotation of the probe beam polarization, which is proportional to the out-of-plane component

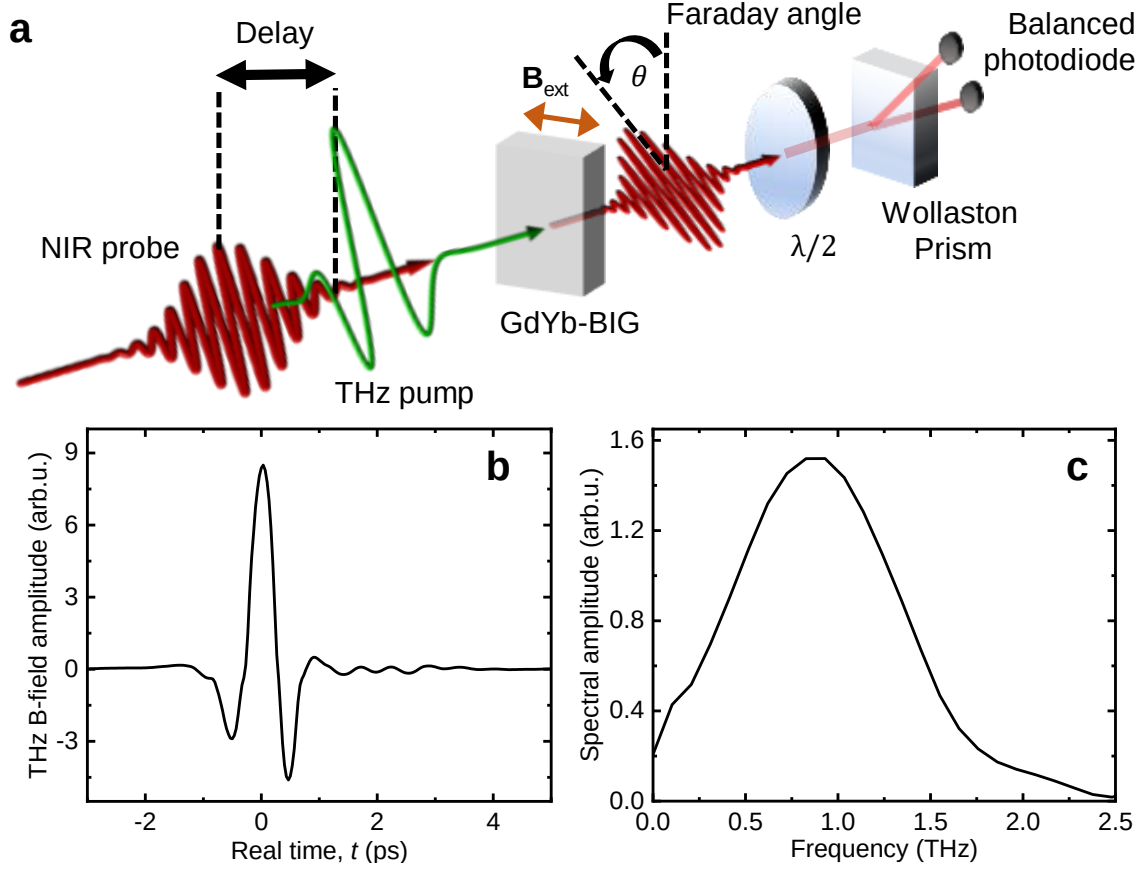


Figure 3.2: **(a)** Schematic illustration of the THz-pump near-infrared (NIR) probe experiment performed in the Faraday geometry. **(b)** Temporal profile of the incident single-cycle THz pulse used to excite the spin dynamics. **(c)** Corresponding Fourier spectrum of the incident THz pulse.

of the magnetization along the probe propagation direction, is measured as a function of the pump-probe delay time using a half-wave plate, Wollaston prism, and balanced photodiode detection scheme, see figure 3.2a. The pump-probe delay is scanned by varying the arrival time of the THz pulse relative to the NIR probe pulse, yielding a time-resolved trace of the magnetization dynamics following THz excitation. All measurements are performed at room temperature in an inert nitrogen atmosphere. Control measurements in the absence of the external magnetic field are also performed to confirm that the observed oscillatory signal is of purely magnetic origin, since as demonstrated in Chapter 4 – the Kaplan-Kittel

exchange mode is only driven when the THz magnetic field has a component transverse to the equilibrium magnetization direction established by the external field. The incident THz pulse and spectra are shown in Figure 3.2b,c, respectively.

3.3 THz time-domain spectroscopy

Terahertz time-domain spectroscopy (THz-TDS) is one of the most widely used techniques in modern condensed matter physics for probing low-energy excitations in materials. Occupying the spectral region between microwave and mid-infrared radiation, THz radiation spans frequencies from approximately 0.1 THz to 10 THz [90], corresponding to photon energies of 0.4-41 meV and wavelengths of 30 μm to 3 mm. This frequency window is particularly well suited to the investigation of collective low-energy excitations in correlated electron systems, including phonons [91, 92], magnons [93, 94], etc., all of which possess characteristic energy scales resonant with THz radiation. In the context of this thesis, THz-TDS serves a dual role: it characterizes the frequency-domain properties of the Kaplan-Kittel exchange resonance in GdYb-BIG through the free-induction decay (FID) of the transmitted THz field, and it provides the linear response baseline against which the nonlinear 2D-THz measurements of Chapter 4 are compared. Here, the free-induction decay (FID) refers to the coherent oscillatory emission from a resonantly driven spin system that persists after the driving field has passed. When the THz pulse resonantly excites the Kaplan-Kittel exchange mode of GdYb-BIG at 0.48 THz, it coherently displaces the sublattice magnetizations from their equilibrium orientations. Once the driving pulse has transmitted through the crystal, the spin system continues to precess freely at its natural resonance frequency in the absence of the driving field, while the magnetization relaxes back toward equilibrium through Gilbert damping. This decaying coherent oscillation appeared as a damped oscillatory tail in the transmitted THz transient. The Fourier transform

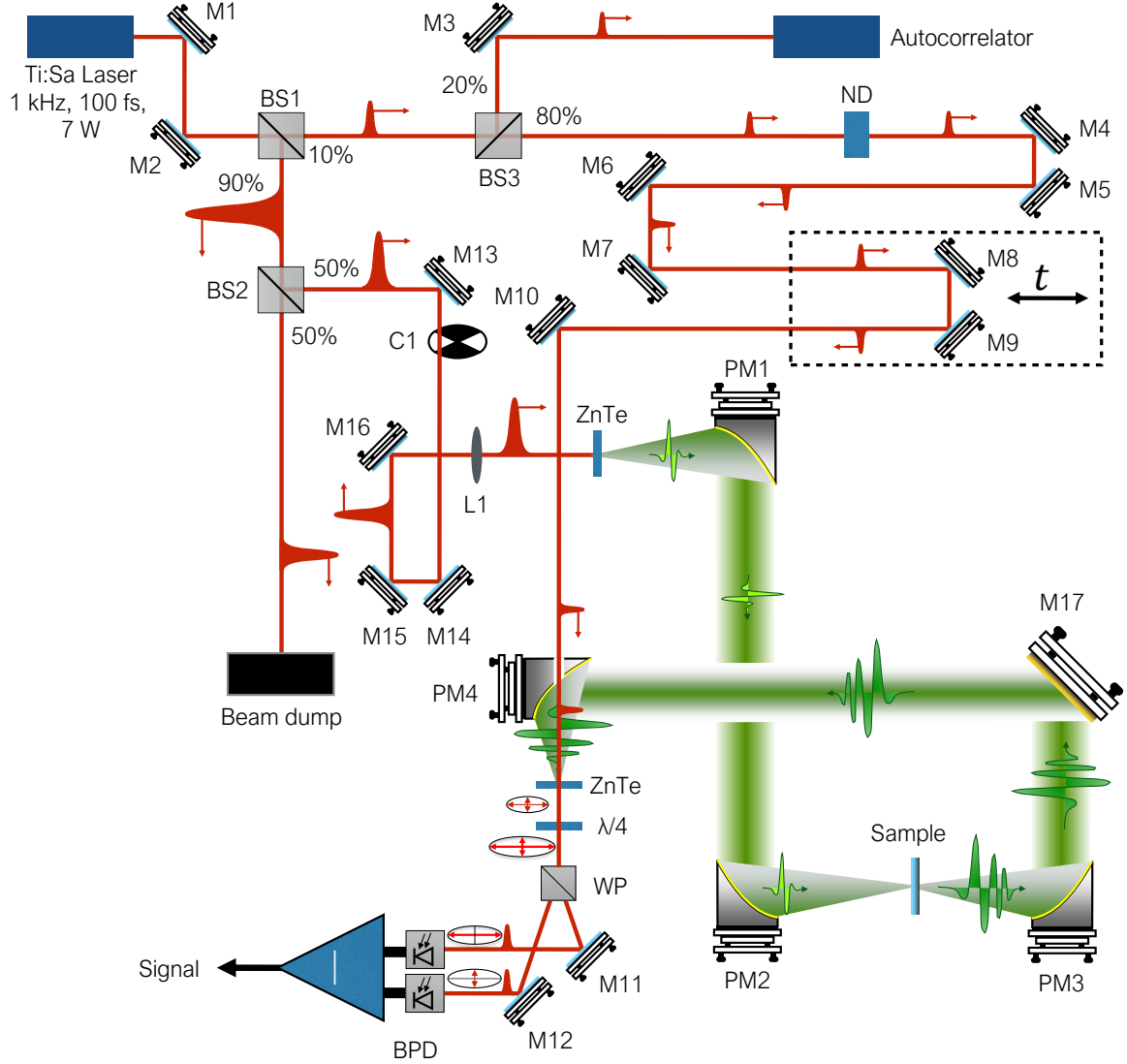


Figure 3.3: Experimental setup for terahertz time-domain spectroscopy (THz-TDS). M: mirror; BS: beam splitter; PM: off-axis parabolic mirror; $\lambda/4$: quarter-wave plate; BPD: balanced photodiode; ND: neutral density filter; and C: optical chopper.

of this tail yields the resonance frequency of the exchange mode, making the FID signal the primary spectroscopic observable in our THz-TDS measurements.

3.3.1 Experimental setup

THz pulses are generated and detected using a standard optical pump-probe geometry driven by a Ti:Sapphire laser operating at 800 nm central wavelength, with 100 fs pulse duration, 1 kHz repetition rate, and 2 mJ pulse energy. The fundamental beam is split into two parts: 90% is used for THz generation and 10% serves as the gating beam for electro-optic detection. Single-cycle THz pulses are generated by optical rectification in a 0.5 mm thick (110)-oriented ZnTe crystal. Detection is performed by free-space electro-optic sampling using a composite ZnTe detection crystal consisting of a 0.5 mm thick (110)-oriented crystal optically bonded to a 2 mm thick (100)-oriented crystal. The THz-induced birefringence in the detection crystal rotates the polarization of the co-propagating gating beam, which is subsequently analyzed using a quarter-wave plate, a Wollaston prism, and a balanced photodiode pair to retrieve the THz electric field transient; see Figure 3.3.

3.3.2 THz generation: Optical rectification

Single-cycle THz pulses are generated by optical rectification [95–97], a second-order nonlinear optical process that occurs in non-centrosymmetric crystals. When an intense femtosecond laser pulse propagates through such a crystal, the nonlinear polarization induced in the medium can be expressed as a power series in the driving electric field E [98],

$$P = \epsilon_0 \left(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots \right), \quad (3.2)$$

where ϵ_0 is the permittivity of free space and $\chi^{(n)}$ is the n -th order electric susceptibility. The first term describes the linear optical response; the higher-order terms govern the nonlinear processes. Optical rectification arises from the $\chi^{(2)}$ term, in which two frequency components ω_1 and ω_2 present within the broadband femtosecond pulse interact to generate a difference-frequency polarization at $\omega_1 - \omega_2$. For the spectrally broad pulse used here, this intra-pulse difference-frequency generation produces a low-frequency polarization that

spans the THz range; see Figure 3.4a. The second-order nonlinear polarization can be written as

$$P_{\text{NL}}^{(2)} = \epsilon_0 \chi^{(2)} \left[2E_0^2 + E_0^2 \cos(2\omega_1 t) + E_0^2 \cos(2\omega_2 t) + E_0^2 \cos((\omega_1 + \omega_2)t) + E_0^2 \cos((\omega_1 - \omega_2)t) \right], \quad (3.3)$$

which contains a DC rectification term (the first term), second-harmonic components at $2\omega_1$ and $2\omega_2$, a sum-frequency term at $\omega_1 + \omega_2$, and the difference-frequency term at $\omega_1 - \omega_2$ that gives rise to THz generation [98, 99].

An essential condition for efficient THz generation is phase matching: the group velocity of the optical pump pulse must be equal to the phase velocity of the generated THz radiation within the crystal, so that THz fields generated at different positions along the propagation direction add constructively. The distance over which this constructive interference is maintained is known as the coherence length, and it determines the optimal crystal thickness for a given pump wavelength and crystal orientation [100, 101]. For 800 nm pulses in (110)-oriented ZnTe, the phase-matching condition is well satisfied over the 0.5 mm crystal length used here, producing THz pulses with spectral content extending to approximately 3 THz.

3.3.3 THz detection: Electro-optic sampling

The THz electric field transient is detected by free-space electro-optic (EO) sampling, based on the linear electro-optic effect (Pockels effect) [102]. When the THz electric field E_{THz} is co-propagating with the gating pulse inside the EO detection crystal, it induces a birefringence proportional to the instantaneous THz field amplitude, modifying the refractive index as

$$\Delta n = \frac{1}{2} n_0^3 r E_{\text{THz}}, \quad (3.4)$$

where n_0 is the unperturbed refractive index and r is the effective EO coefficient for the

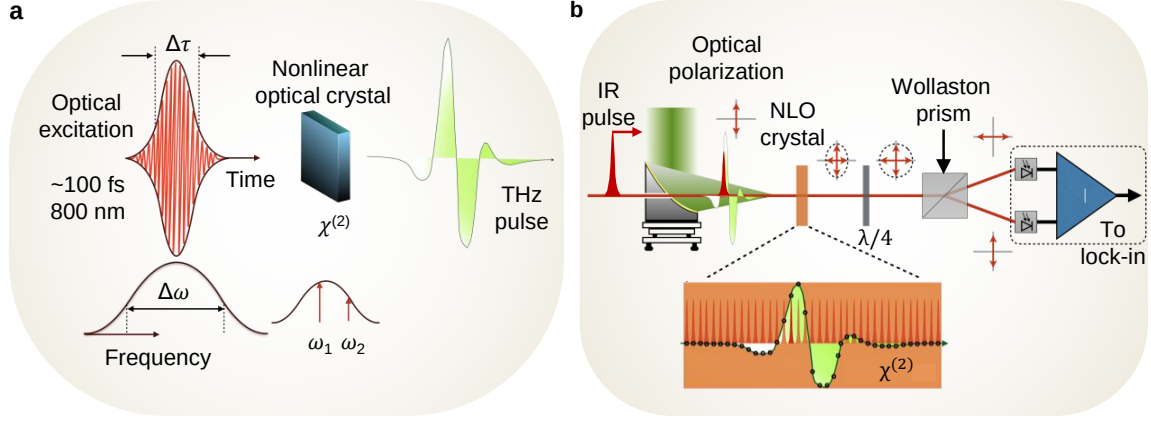


Figure 3.4: **(a)** Schematic illustration of terahertz pulse generation via optical rectification in a non-centrosymmetric crystal. **(b)** Schematic representation of the electro-optic sampling technique used for detection of the generated THz pulse.

chosen crystal geometry and orientation. This field-induced birefringence rotates the polarization of the co-propagating gating beam by an amount proportional to E_{THz} . After passing through a quarter-wave plate, which converts the elliptical polarization induced by the birefringence into a differential intensity signal between two orthogonal linear components, a Wollaston prism separates the two components, which are measured by a balanced photodiode. The resulting difference signal ΔI_p , normalized by the total probe intensity I_p , is directly proportional to the THz electric field: [100]

$$E_{\text{THz}} = \frac{\Delta I_p}{I_p} \cdot \frac{\lambda_L}{2\pi L r n_0^3}, \quad (3.5)$$

where λ_L is the probe wavelength and L is the crystal thickness. By scanning the arrival time of the gating pulse relative to the THz pulse using a mechanical delay stage, the full time-domain THz waveform is reconstructed stroboscopically, point by point; see Figure 3.4b. A Fourier transform of the resulting time-domain transient then yields the amplitude and phase spectra of the THz field over the full detection bandwidth, from which the complex optical response, such as refractive index, absorption coefficient, and conductivity of the

sample, can be extracted.

3.4 Nonlinear two-dimensional THz spectroscopy

The linear THz-TDS technique described in the preceding section accesses only the first-order response of the spin system - that is, the regime in which the magnetization deviation from equilibrium scales linearly with the driving magnetic field amplitude. While this is sufficient to identify and characterize the exchange resonance mode of GdYb-BIG, it is fundamentally insensitive to the higher-order interaction pathways that govern the nonlinear spin dynamics investigated in Chapter 4. Accessing the nonlinear regime requires a technique that can simultaneously excite and resolve multiple field-spin interaction pathways in both the time and frequency domains, provided by nonlinear two-dimensional THz spectroscopy (2D-THz spectroscopy) [102, 103]. This technique, the central experimental tool of this thesis, extends the single-pulse THz-TDS scheme to a dual-pulse geometry and provides direct, phase-resolved access to the $\chi^{(3)}$ nonlinear spin dynamics.

3.4.1 Experimental Setup

The 2D THz spectroscopy measurements are performed using two collinear, co-propagating THz pulses A and B, separated by a controllable time delay τ . The 90% portion of the fundamental Ti:Sapphire beam, carrying 6 mJ pulse energy, is divided into two equal parts for the generation of the two THz pulses. The first THz pulse (B) is generated by optical rectification in the 0.5 mm thick (110)-oriented ZnTe crystal described in the preceding section. The second THz pulse (A) is generated independently by optical rectification in a 0.5 mm thick (110)-oriented GaP crystal. Using a second motorized delay stage, the arrival time of pulse A on the sample is varied systematically with respect to pulse B, which arrives at a fixed time, see Figure 3.5. The two pulses interact with the sample collinearly, so that all nonlinear signals generated by their combined interaction are simultaneously encoded

in the transmitted field and can be retrieved in a single 2D scan without phase-matching constraints. The nonlinear THz field is extracted by subtracting the individual single-pulse responses from the total two-pulse response:

$$\mathbf{E}_{\text{NL}}(t, \tau) = \mathbf{E}_{\text{AB}}(t, \tau) - \mathbf{E}_{\text{A}}(t, \tau) - \mathbf{E}_{\text{B}}(t), \quad (3.6)$$

where $\mathbf{E}_{\text{AB}}(t, \tau)$ is the transmitted field when both pulses are present, and $\mathbf{E}_{\text{A}}(t, \tau)$, $\mathbf{E}_{\text{B}}(t)$ are the transmitted fields with only pulse A or B present, respectively. This field subtraction, made possible by the direct field detection of THz radiation via electro-optic sampling, eliminates all contributions that scale linearly with either pulse individually, leaving only the cross-terms that arise from the nonlinear interaction of the two pulses with the system.

3.4.2 Frequency-vector analysis of nonlinear signals

The phase fronts of the two THz pulses in the (t, τ) plane, illustrated in Figure 3.6a, determine the allowed frequency vectors ν_{A} (green arrow) and ν_{B} (red arrow) and therefore the positions at which the nonlinear signals appear in the 2D map. Pulse A has a phase front tilted at 45° in the (t, τ) plane, because it depends on both the real time t and the delay τ , while pulse B has a vertical phase front since it arrives at a fixed time independent of τ . This geometric difference directly determines the frequency-vector directions and the resulting 2D spectral pattern.

The positions of the nonlinear signals in the 2D frequency map can be systematically identified using the frequency-vector formalism, in which each field interaction between a THz pulse and the spin system is represented by the frequency vector of the corresponding pulse, with a positive or negative sign indicating whether that interaction contributes a positive or negative phase to the overall nonlinear pathway. For a third-order, $\chi^{(3)}$, process involving three field interactions at the collective mode frequency ν_0 , the emitted nonlinear signal must satisfy $\pm\nu_0 \pm \nu_0 \pm \nu_0 = \nu_{\text{signal}}$, where two of the three interactions are provided

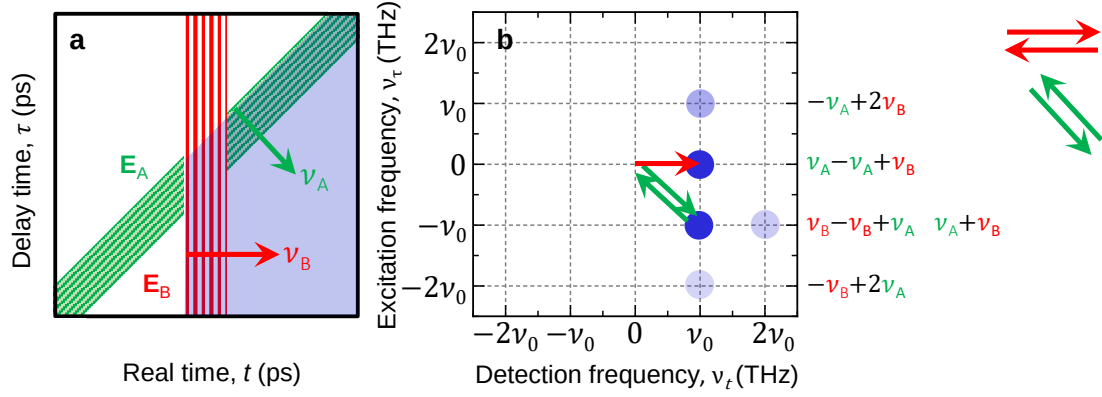


Figure 3.6: **(a)** Electric-field profiles of the two incident THz pulses, A and B, plotted as a function of the real time (t) and the inter-pulse delay time (τ). The corresponding frequency vectors of pulses A and B are indicated by the green (ν_A) and red (ν_B) arrows, respectively. **(b)** Nonlinear 2D-THz spectra plotted as a function of the detection frequency (ν_t) and the excitation frequency (ν_τ).

and provides the emitted signal frequency ν_0 . This interaction sequence is schematically illustrated in Figure 3.6b. Similarly, the photon-echo signal at $(\nu_0, +\nu_0)$ arises from the sequence $2\nu_B - \nu_A$, in which pulse B interacts twice with the same phase and pulse A interacts once with the opposite phase, such that the phase information from both pulses is retained in the emitted signal, the defining property that makes echo signals sensitive to the decoherence timescale of the collective mode.

3.4.3 Physical picture: Three regimes of two-pulse interaction

The physical picture of a 2D-THz measurement can be understood by considering three distinct regimes of the inter-pulse delay τ relative to two characteristic timescales of the system: the individual pulse duration Γ and the lifetime of the excited collective mode t_{exc} .

In the first regime, $\tau < \Gamma$, the two pulses overlap temporally and the sample cannot resolve them as separate excitations. The system effectively experiences a single enhanced field $\mathbf{E}_A + \mathbf{E}_B$, and the nonlinear response is dominated by higher-harmonic generation from the combined field. In the 2D frequency map, this regime produces signals at the

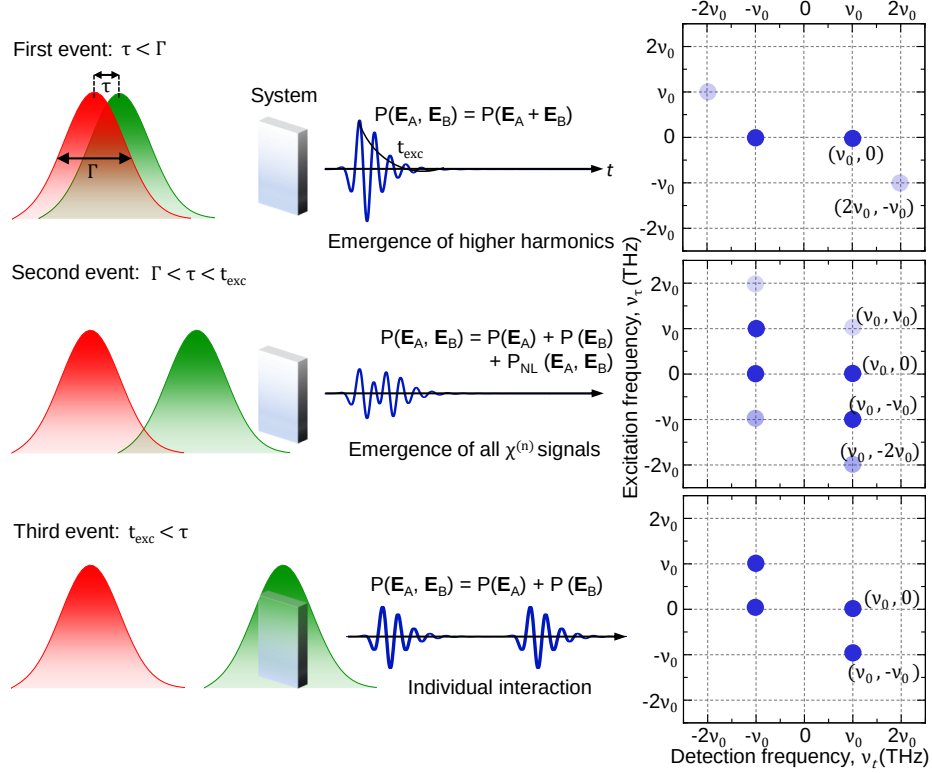


Figure 3.7: Schematic representation of the interaction of two time-delayed THz pulses with the sample. The complete process is divided into three interaction events. The green and red Gaussian envelopes represent the incident THz electric fields $\mathbf{E}_A(t, \tau)$ and $\mathbf{E}_B(t)$, respectively, separated by a temporal delay τ . Here, Γ denotes the pulse duration of the individual THz pulses, while t_{exc} represents the lifetime of the excited collective mode at the resonance frequency ν_0 . The blue curve illustrates a representative temporal scan of the emitted THz field during the three interaction events. The corresponding nonlinear responses are shown on the right in the two-dimensional frequency domain as a function of the detection frequency (ν_t) and the excitation frequency (ν_τ). The darker and lighter shades of blue indicate the relative intensities of the nonlinear signals.

fundamental mode frequency $(\nu_0, 0)$ and its second harmonic $(2\nu_0, -\nu_0)$, as illustrated in Figure 3.7 top panel.

In the second regime, $\Gamma < \tau < t_{\text{exc}}$, the pulses are temporally separated but the collective mode excited by the first pulse has not yet decayed when the second pulse arrives. The system therefore retains a memory of the first interaction, and its nonlinear susceptibility is transiently modified as $\chi^{(n)} = \chi_0^{(n)} + \Delta\chi^{(n)}$, where $\Delta\chi^{(n)}$ encodes the change induced

by the first pulse [36, 104]. The second pulse then interacts with this modified susceptibility, giving rise to a set of nonlinear signals in the 2D map, including pump-probe signals at $(\nu_0, 0)$ and $(\nu_0, -\nu_0)$, and photon-echo signals at $(\nu_0, +\nu_0)$ and $(\nu_0, -2\nu_0)$, as shown in Figure 3.7 middle panel.

In the third regime, $\tau > t_{\text{exc}}$, the collective mode has fully decayed before the second pulse arrives, and the two pulses interact with the sample independently. The 2D response reduces to two superimposed single-pulse responses, and no cross-nonlinearities are observable, as shown in Figure 3.7 bottom panel. Since the total two-pulse response $\mathbf{E}_{\text{AB}}(t, \tau)$ contains both the nonlinear cross-terms and the individual single-pulse contributions, the nonlinear signal is isolated by performing a field subtraction directly in the time domain, as defined in Equation (3.6). This subtraction is made possible by the coherent, phase-sensitive nature of electro-optic detection, which measures the THz electric field amplitude directly rather than its intensity, which is unique to THz spectroscopy and has no direct analogue in conventional nonlinear optical techniques such as second- or third-harmonic generation.

Chapter 4

Evidence of relativistic field derivative torque in nonlinear THz response of magnetization dynamics

Having established the theoretical framework governing ultrafast spin dynamics in the preceding chapters, the question naturally emerges: is the conventional LLG equation truly complete for describing THz-driven spin dynamics in systems with high-frequency collective excitations and appreciable Gilbert damping, or do relativistic corrections become indispensable? As discussed in Chapter 2, the answer lies in the field-derivative torque - a relativistic correction arising from the spin-orbit coupling of the system, which predicts that not only the magnetic field component of the THz pulse, but also its time derivative, exerts a torque on the spin system [22, 44]. While the FDT has been theoretically proposed and studied for over a decade, its direct experimental realization has remained elusive. Prior attempts to probe its role in ferrimagnetic systems were hindered by two key material limitations: a relatively low Gilbert damping parameter and inequivalent sublattice gyromagnetic ratios, both of which suppress the FDT contribution below experimentally resolvable levels [20].

These limitations underscore the need for a material system in which the conditions are simultaneously favorable for hosting a THz-frequency exchange resonance, sustaining a comparatively large Gilbert damping, and maintaining equivalent sublattice gyromagnetic ratios - precisely the combination required for the FDT contribution to become significant and isolable. The rare-earth iron garnet $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ satisfies all of these conditions, making it a uniquely suitable platform for the experimental investigation of relativis-

tic spin torques. The present chapter reports the experimental realization of this objective: using linear THz spectroscopy, THz-pump optical-probe Faraday rotation, and nonlinear two-dimensional THz spectroscopy, we demonstrate that the observed nonlinear magnetization dynamics of GdYb-BIG cannot be described by the conventional Zeeman torque alone, and that incorporating the FDT into the LLG framework is essential to reproduce the experimental observations.

4.1 Material: $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$

The material investigated throughout this chapter is a single crystal of the rare-earth bismuth-doped iron garnet $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$, hereafter referred to as GdYb-BIG. The crystals were grown by Prof. Takuya Satoh using the liquid-phase epitaxy method, oriented along the [111] crystallographic direction, and have a thickness of $380\text{ }\mu\text{m}$. The material orders ferrimagnetically in its ground state with a Curie temperature of 573 K and a magnetization compensation temperature of $T_{\text{MC}} = 96\text{ K}$.

The structural basis for the ferrimagnetic order is illustrated in Figure 4.1a. The rare-earth ions Gd^{3+} and Yb^{3+} , residing at the dodecahedral sites, couple antiferromagnetically to the net magnetization of the Fe^{3+} ions occupying the tetrahedral and octahedral sites, via superexchange interaction, mediated by the nonmagnetic O^{2-} ions. The system can thus be effectively modeled as a two-sublattice ferrimagnet with unequal rare-earth and iron sublattice magnetizations [1, 45, 47]. The strong inter-sublattice exchange interaction between these two magnetic sublattices gives rise to high-frequency collective spin excitations in the THz regime, whose character and frequencies can be understood from the equations of motion of the two-sublattice system.

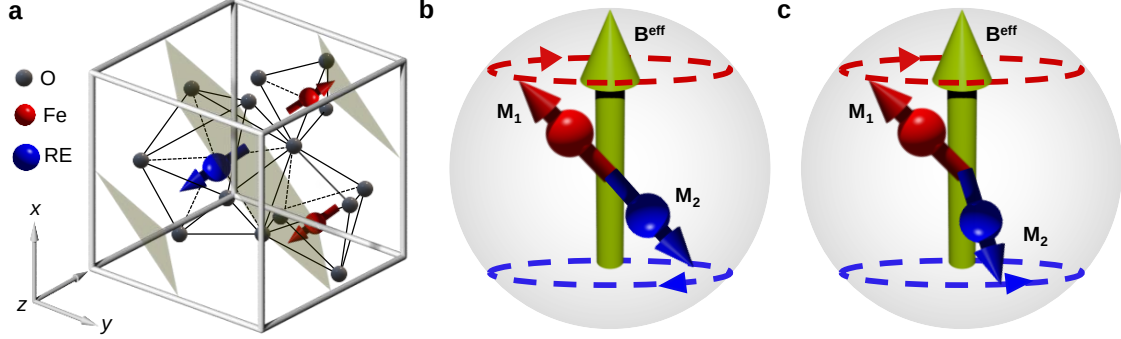


Figure 4.1: **(a)** Crystal structure of the GdYb-BIG unit cell, illustrating the arrangement of the rare-earth and Fe sublattices. **(b)** Low-frequency backward volume magnetostatic wave (BVMW) mode, in which the two sublattice magnetizations precess nearly in phase. **(c)** High-frequency Kaplan-Kittel exchange mode, characterized by the out-of-phase precession of the rare-earth and Fe sublattice magnetizations driven by the strong inter-sublattice exchange interaction.

4.1.1 Kaplan-Kittel exchange resonance mode

A quantitative understanding of the collective spin dynamics in a two-sublattice ferrimagnet follows from the analysis of the equations of motion for each sublattice magnetization in the presence of an applied external magnetic field $\mathbf{H}_{\text{ext}}$. In general, the number of distinct self-modes of oscillation is equal to the number of magnetic sublattices; for the simplest case of a two-sublattice ferrimagnet, neglecting anisotropy and damping, the equations of motion for the two sublattice magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$ take the form

$$\frac{d\mathbf{M}_1}{dt} = \gamma_1 \mathbf{M}_1 \times (\mathbf{H}_{\text{ext}} - \mathbf{H}_{\text{ex}}), \quad \frac{d\mathbf{M}_2}{dt} = \gamma_2 \mathbf{M}_2 \times (\mathbf{H}_{\text{ext}} - \mathbf{H}_{\text{ex}}), \quad (4.1)$$

where γ_1 and γ_2 are the sublattice gyromagnetic ratios and $\mathbf{H}_{\text{ex}}$ is the inter-sublattice exchange field, defined as

$$\mathbf{H}_{\text{ex}} = -N_{12} (\mathbf{M}_1 + \mathbf{M}_2), \quad (4.2)$$

with N_{12} being the molecular field coefficient coupling the two sublattices. Two distinct normal modes emerge from this system of equations, corresponding to two types of collec-

tive spin precession. The first is a ferromagnetic-type mode, in which the two sublattice magnetizations precess in phase with one another, at a frequency determined primarily by the external and anisotropy fields,

$$\omega_{\text{FM}} = \gamma_{\text{eff}} H_{\text{eff}}, \quad (4.3)$$

where γ_{eff} and H_{eff} denote the effective gyromagnetic ratio and effective field of the combined system, respectively. The dynamics of a two-sublattice ferrimagnet can be described in terms of the net magnetization and the relative orientation of the two sublattices. For the two collective modes, the effective gyromagnetic ratio and effective magnetic field are defined as

$$\gamma_{\text{eff}} = \frac{\gamma_1 M_1 - \gamma_2 M_2}{M_1 - M_2}, \quad (4.4)$$

and

$$H_{\text{eff}} = H_{\text{ext}} + H_{\text{ani}} + H_{\text{dem}}, \quad (4.5)$$

where γ_1 and γ_2 are the gyromagnetic ratios and M_1 and M_2 are the magnitudes of the two sublattice magnetizations. Here, H_{ext} , H_{ani} , and H_{dem} denote the external, anisotropy, and demagnetization fields, respectively. When $\gamma_1 = \gamma_2 = \gamma$, the effective gyromagnetic ratio reduces to $\gamma_{\text{eff}} = \gamma$. The two-sublattice equations of motion give two characteristic modes. The first is a ferromagnetic-type mode, in which the two sublattice magnetizations precess predominantly in phase. In GdYb-BIG, this mode corresponds to the low-frequency backward-volume magnetostatic wave (BVMW) mode at approximately 4.2 GHz in the presence of an in-plane field of 120 mT. The second is an antiferromagnetic-type mode, known as the Kaplan-Kittel exchange mode, in which the two sublattice magnetizations precess predominantly out of phase. In the limit where the inter-sublattice exchange field is much larger than the applied and anisotropy fields, the frequency of this exchange mode is given by

$$\omega_{\text{KK}} = \sqrt{\gamma_1 \gamma_2} N_{12} (M_2 - M_1), \quad (4.6)$$

where N_{12} is the inter-sublattice exchange parameter and M_1 and M_2 are the magnitudes of the two sublattice magnetizations. This is the exchange-dominated Kaplan-Kittel limit of the two-sublattice dynamics.

Since the exchange field is several orders of magnitude larger than the applied and anisotropy fields, the Kaplan-Kittel mode occurs at a frequency far exceeding that of the ferromagnetic-type mode, placing it in the THz frequency range. For GdYb-BIG, the experimentally observed exchange resonance occurs at 0.48 THz. The value of the exchange parameter used in the numerical simulations is chosen such that the coupled LLG equations reproduce this experimentally observed resonance frequency.

It is also important to note that in GdYb-BIG, the Fe^{3+} and the rare-earth sublattices have been reported to possess equal gyromagnetic ratios, $\gamma_{\text{Fe}} = \gamma_{\text{RE}} = 1.76 \times 10^{11} \text{ s}^{-1}\text{T}^{-1}$ [1]. This condition requires particular consideration in rare-earth iron garnets, where the gyromagnetic ratios of the two sublattices can generally differ because of the contribution of orbital angular momentum. At room temperature, the rare-earth contribution to the magnetization of GdYb-BIG is dominated by the Gd^{3+} ions. The Fe^{3+} ions occupy octahedral and tetrahedral crystal sites, where the crystal field from the surrounding oxygen ions largely quenches the orbital magnetic moment. Consequently, the magnetic moment of Fe^{3+} is predominantly spin-derived, with $g \approx 2$. In contrast, the $4f$ electrons of rare-earth ions are strongly shielded by the outer $5s$ and $5p$ orbitals, so that the crystal field does not fully quench the orbital angular momentum. As a result, both spin and orbital angular momenta can contribute through spin-orbit coupling, generally leading to g -factors different from 2. However, for Gd^{3+} , the orbital angular momentum is zero ($L = 0$), and its magnetic moment is predominantly spin-derived, giving $g \approx 2$, similar to Fe^{3+} . Therefore, at room temperature, the gyromagnetic ratios of the Fe and rare-earth (Gd) sublattices in GdYb-BIG can be considered approximately equivalent. This equivalent gyromagnetic ratio is also important for the observation of the field-derivative torque, as discussed in Chapter 5. Deviations

between the gyromagnetic ratios of the two sublattices can cause the conventional Zeeman-torque contribution to dominate over the FDT contribution. Together with the relatively large Gilbert damping ($\alpha \approx 0.02$) and the THz-frequency Kaplan-Kittel exchange mode, the equivalent sublattice gyromagnetic ratios make GdYb-BIG a suitable material system for observing the FDT experimentally. With the material system and its characteristic spin dynamics established, we now turn to the experimental identification and characterization of the Kaplan-Kittel mode through THz-pump optical-probe and linear THz time-domain spectroscopy measurements.

4.2 Identifying the exchange resonance mode: THz-pump Optical-probe measurement

With the material platform established, the first experimental step is to identify the high-frequency exchange resonance mode of GdYb-BIG and confirm its resonant excitation by the THz pulse. To this end, we perform a THz-pump optical-probe experiment in the Faraday geometry. In this measurement scheme, an intense single-cycle THz pulse drives the magnetization dynamics in the sample, which are subsequently probed by a co-propagating near-infrared (NIR) pulse via the magneto-optical Faraday effect. The rotation of the NIR probe polarization after passing through the sample is recorded as a function of pump-probe delay time, providing a direct time-resolved trace of the out-of-plane magnetization component induced by the THz excitation. The measurements are carried out both in the presence and in the absence of an in-plane external static magnetic field.

As shown in Figure 4.2a, a clear and sustained oscillatory modulation of the Faraday rotation is observed as a function of pump-probe delay time when an in-plane external magnetic field is applied. These oscillations are completely absent when the external magnetic field is removed, confirming that the observed dynamics are of purely magnetic origin and are governed by the field orientation – consistent with the symmetry requirements of

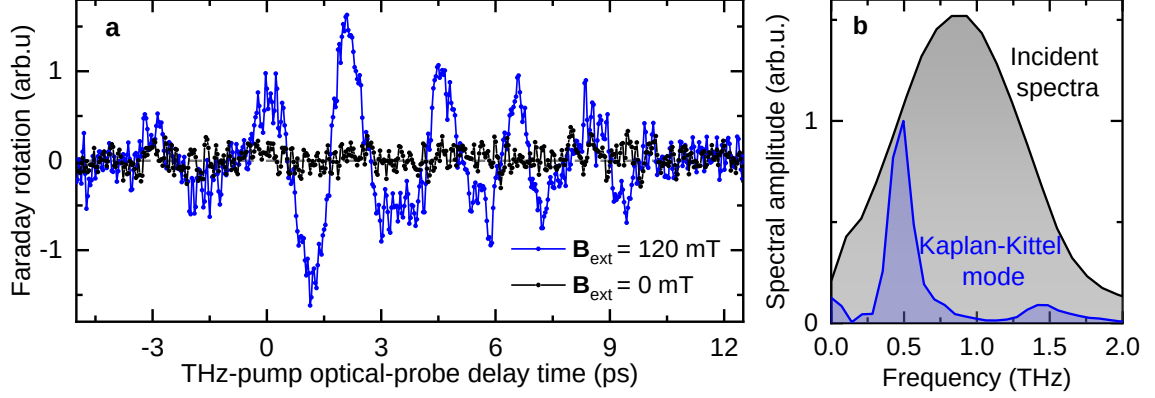


Figure 4.2: **(a)** Time-resolved Faraday rotation of GdYb-BIG at room temperature as a function of the THz-pump NIR-probe delay, measured in the presence (blue) and absence (black) of an in-plane external magnetic field. The oscillatory response observed under the applied magnetic field arises from the coherent excitation of the exchange mode by the incident THz pulse. **(b)** Normalized Fourier transform of the time-domain signal shown in (a), revealing a pronounced resonance at 0.48 THz corresponding to the Kaplan-Kittel exchange mode. The black curve represents the spectrum of the incident THz pulse used for excitation.

the Kaplan-Kittel exchange mode, where the THz magnetic field must have a component transverse to the equilibrium magnetization in order to exert the Zeeman torque that drives the anti-phase sublattice precession. The corresponding frequency spectrum, obtained by performing FFT of the time-domain signal in Figure 4.2a, is presented in Figure 4.2b. A single sharp resonance peak appears at 0.48 THz, confirming the resonant excitation of the Kaplan-Kittel exchange mode by the THz pulse. These results establish the presence of the exchange mode in GdYb-BIG and motivate a more detailed characterization through direct linear THz transmission measurements, as discussed in the following section.

4.3 Characteristics of the exchange mode: Linear THz-TDS

To further characterize the exchange resonance mode and access its spectral properties directly, we perform linear THz time-domain spectroscopy (THz-TDS) in the transmission

geometry on the GdYb-BIG sample. In this experiment, the time-varying magnetic field of the incident THz pulse exerts a Zeeman torque on the magnetic moments of both sublattices, modifying the sublattice-dependent effective magnetic field. This perturbation drives the ferrimagnetic system out of equilibrium and coherently excites the exchange mode. Following the excitation, the magnetization relaxes back to its equilibrium configuration via a free-induction decay (FID), which is directly encoded in the transmitted THz signal and serves as a spectroscopic fingerprint of the exchange resonance.

The transmitted THz time-domain signals are recorded in the absence and in the presence of an external magnetic field $\mathbf{B}_{\text{ext}} = 120 \text{ mT}$, applied in two distinct orientations with respect to the incident THz magnetic field $\mathbf{B}_{\text{THz}}$: a parallel orientation ($\mathbf{B}_{\parallel}$) and a perpendicular orientation ($\mathbf{B}_{\perp}$), as illustrated schematically in Figure 4.3a. Reference measurements are performed in free space without the sample to establish the incident field profile. The amplitude of the THz signal transmitted through GdYb-BIG is significantly attenuated compared to the incident field, reflecting the strong absorption of the material. In addition, the transmitted pulse is delayed by approximately 5.76 ps relative to the reference, a shift that arises from the combined effect of the sample thickness and its high refractive index. From the $\mathbf{B}_{\text{ext}} = 0 \text{ mT}$ measurement, we extract a refractive index of approximately 5 and an absorption coefficient reaching up to 20 cm^{-1} across a broad THz frequency bandwidth, shown in Figures 4.3c and 4.3d. The strong broadband absorption results in a heavily convoluted FID signal, making it difficult to isolate the narrow-band exchange resonance directly from the raw time-domain trace.

To circumvent this, the spectra measured under an applied external magnetic field are normalized by the spectrum recorded at $\mathbf{B}_{\text{ext}} = 0 \text{ mT}$, thereby suppressing the dominant non-magnetic absorption background and exposing any field-induced changes in the transmission. As shown in the inset of Figure 4.3b, the normalized transmittance under the parallel field configuration ($\mathbf{B}_{\parallel}$) is nearly identical to the zero-field reference, indicating

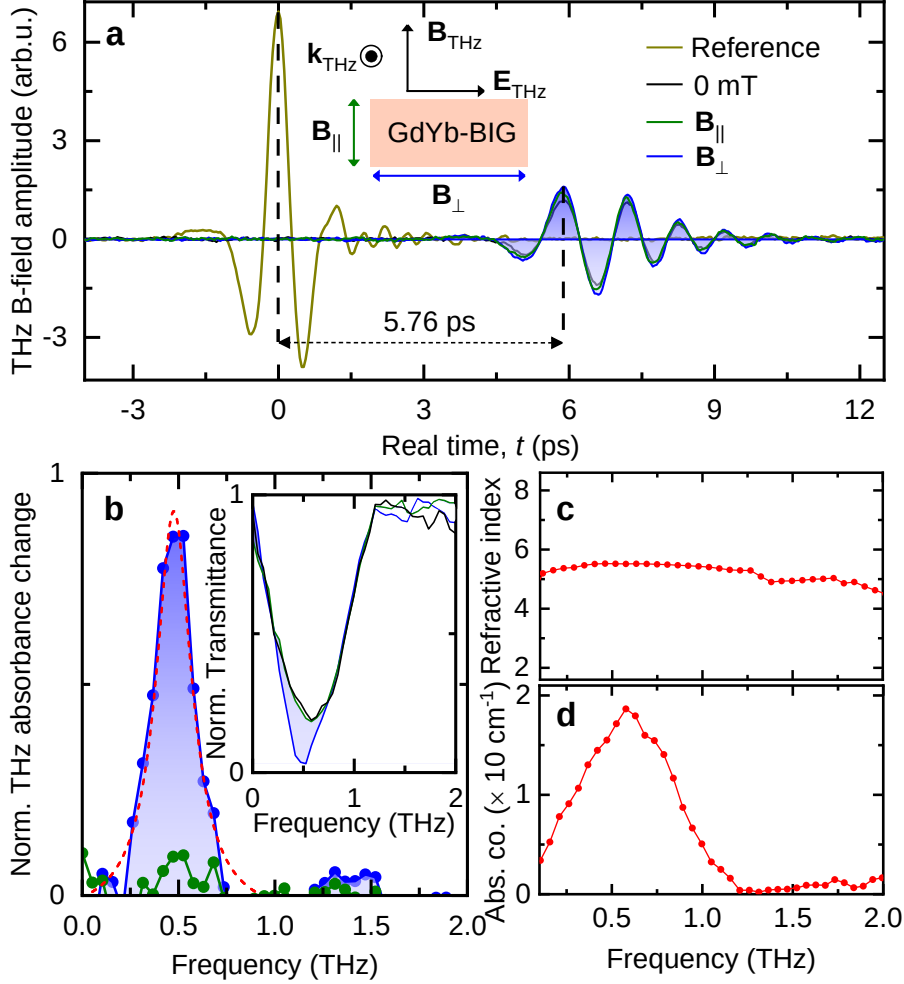


Figure 4.3: **(a)** Time-domain THz magnetic field transients measured for the reference and the GdYb-BIG sample under different orientations of the external magnetic field with respect to the incident THz magnetic field. **(b)** Normalized absorbance change showing the Kaplan-Kittel exchange resonance at 0.48 THz. The red dashed line represents the Lorentzian fit to the experimental data. The inset displays the corresponding normalized transmittance spectra for different magnetic field orientations. **(c)** Frequency-dependent refractive index and **(d)** absorption coefficient of GdYb-BIG, measured in the absence of an external static magnetic field.

that this orientation does not couple to the exchange mode. In sharp contrast, when the external magnetic field is applied perpendicular to $\mathbf{B}_{\text{THz}}$ (i.e., the $\mathbf{B}_{\perp}$ configuration), a pronounced additional absorption dip appears at 0.48 THz, giving rise to a well-defined peak in the normalized absorbance spectrum shown in Figure 4.3b. This feature is well described

by a Lorentzian lineshape centered at 0.48 THz and corresponds directly to the Kaplan-Kittel exchange resonance mode identified in the Faraday measurements of the preceding section. The agreement between these two complementary measurements, time-domain Faraday rotation and frequency-domain THz transmission, provides unambiguous spectroscopic identification of the exchange resonance mode at 0.48 THz in GdYb-BIG. With the exchange mode firmly established, the following sections turn to its dynamical modeling within the LLG framework and, subsequently, to the nonlinear THz response that reveals the indispensable role of the field-derivative torque.

4.4 Theoretical modeling of the exchange mode dynamics

Having experimentally identified the Kaplan-Kittel exchange mode at 0.48 THz through both THz-pump optical-probe and linear THz-TDS measurements, we now turn to its theoretical description within the framework of the coupled Landau-Lifshitz-Gilbert equations. This modeling serves two purposes: first, to confirm that the two coexisting precession modes of GdYb-BIG – the low-frequency BVMW mode and the high-frequency Kaplan-Kittel mode, are correctly reproduced by the conventional LLG framework under impulsive (optical) excitation; and second, to establish the baseline for the correct parameters against which the nonlinear THz response will be compared in the following sections, where the conventional description is shown to be insufficient.

The non-thermal magnetization dynamics are described by the coupled LLG equations for the two-sublattice ferrimagnetic system, introduced in Chapter 2. The time evolution of the sublattice magnetizations is governed by

$$\frac{d\mathbf{M}_i}{dt} = -\frac{\gamma_i}{1 + \alpha_i^2} (\mathbf{M}_i \times \mathbf{B}_i^{\text{eff}}) - \frac{\gamma_i \alpha_i}{(1 + \alpha_i^2)|\mathbf{M}_i|} \mathbf{M}_i \times (\mathbf{M}_i \times \mathbf{B}_i^{\text{eff}}), \quad (4.7)$$

where $\mathbf{M}_i$ ($i = \text{RE, Fe}$) denotes the sublattice magnetization, and γ_i and α_i are the corre-

sponding gyromagnetic ratio and Gilbert damping parameter. The first term on the right-hand side drives the coherent precession of $\mathbf{M}_i$ around the effective magnetic field $\mathbf{B}_i^{\text{eff}}$, while the second term governs the relaxation of the magnetization toward equilibrium at a rate set by α_i . The effective field is obtained from the total free energy density Φ via the functional derivative $\mathbf{B}_i^{\text{eff}} = -\delta\Phi/\delta\mathbf{M}_i$, where Φ incorporates contributions from the inter-sublattice exchange interaction, the uniaxial magnetocrystalline anisotropy, the Zeeman coupling to the external static field, and the demagnetization energy:

$$\begin{aligned} \Phi = & -\lambda \mathbf{M}_{\text{Fe}} \cdot \mathbf{M}_{\text{RE}} - K_{\text{Fe}} \frac{(\mathbf{M}_{\text{Fe}} \cdot \hat{\mathbf{n}})^2}{|\mathbf{M}_{\text{Fe}}|^2} - K_{\text{RE}} \frac{(\mathbf{M}_{\text{RE}} \cdot \hat{\mathbf{n}})^2}{|\mathbf{M}_{\text{RE}}|^2} \\ & - \mu_0 \mathbf{H}_{\text{ext}} \cdot (\mathbf{M}_{\text{Fe}} + \mathbf{M}_{\text{RE}}) + \frac{\mu_0}{2} (\mathbf{M}_{\text{Fe}} \cdot \hat{\mathbf{n}} + \mathbf{M}_{\text{RE}} \cdot \hat{\mathbf{n}})^2, \end{aligned} \quad (4.8)$$

where λ is the inter-sublattice exchange constant, K_{Fe} and K_{RE} are the uniaxial anisotropy constants for the iron and rare-earth sublattices respectively, $\hat{\mathbf{n}}$ is the unit vector along the [111] easy axis, and μ_0 is the permeability of free space. The last term in Equation (4.8) accounts for the demagnetization energy, which in GdYb-BIG is comparable in magnitude to the applied external field and must therefore be retained. The numerical values of all parameters used in the simulations are listed in Table 4.1.

To obtain the ground state of the ferrimagnet, the static external field $\mathbf{B}_{\text{ext}}$ is applied along the in-plane y -direction to saturate the sublattice magnetizations, establishing a well-defined initial configuration from which the THz-induced dynamics can be tracked. Rather than initializing from a purely collinear ferrimagnetic ground state, a slightly canted initial condition is chosen to allow both the low-frequency and high-frequency modes to be captured simultaneously in the relaxation dynamics. The coupled LLG equations are then integrated numerically, and the resulting time evolution is analyzed through the antiferromagnetic Néel vector, $\mathbf{L} = \mathbf{M}_{\text{RE}} - \mathbf{M}_{\text{Fe}}$, which serves as the natural order parameter for tracking the anti-phase sublattice dynamics.

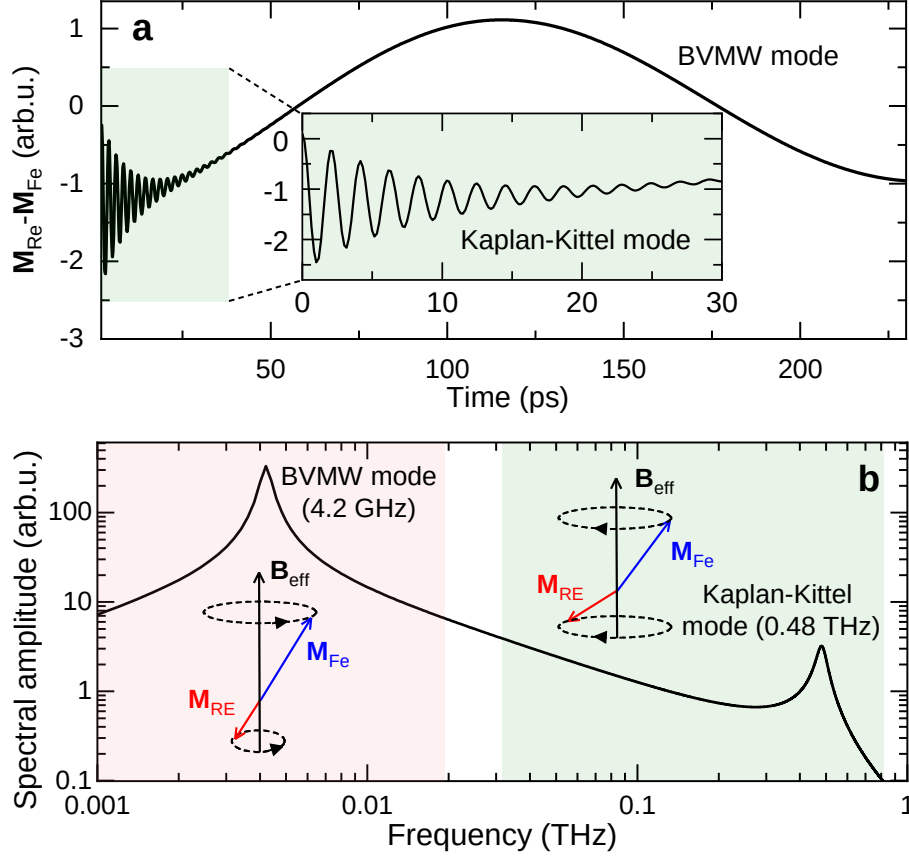


Figure 4.4: **(a)** Simulated time-resolved precession of the antiferromagnetic Néel vector, ($\mathbf{M}_{\text{Re}} - \mathbf{M}_{\text{Fe}}$), about the effective magnetic field, $\mathbf{B}_{\text{eff}}$, obtained using the conventional Landau-Lifshitz-Gilbert (LLG) equation. **(b)** Corresponding Fourier spectrum of the simulated dynamics, revealing a low-frequency backward volume magnetostatic wave (BVMW) mode at 4.2 GHz and a high-frequency Kaplan-Kittel exchange mode at 0.48 THz. The schematics illustrate the relative precessional motion of the rare-earth ($\mathbf{M}_{\text{Re}}$) and transition-metal ($\mathbf{M}_{\text{Fe}}$) sublattice magnetizations associated with the two resonance modes.

The simulations reproduce both precession modes of GdYb-BIG in good agreement with the experimental observations, see Figure 4.4a. The low-frequency BVMW mode appears at 4.2 GHz under an applied field of 120 mT and corresponds to a nearly collinear, in-phase precession of the two sublattice magnetizations around the effective field. The high-frequency Kaplan-Kittel mode appears at 0.48 THz and corresponds to an exchange-driven, anti-phase non-collinear precession of the two sublattices, in which the strong inter-

Table 4.1: Overview of the parameters used in the numerical modelling. Here, K_{Fe} and K_{RE} denote the uniaxial anisotropy constants corresponding to the two sublattices along [111] easy axis direction. The value of the exchange constant (λ) is adjusted to match the Kaplan-Kittel mode obtained in our experiments.

Parameters	Values	References
Exchange constant (λ)	$-1930 \times 10^{-7} \text{ T}^2 \text{ m}^3 / \text{J}$	—
Anisotropy constants ($K_{\text{Fe}} = K_{\text{RE}}$)	1000 J/m^3	Ref. [1]
Gilbert damping parameter ($\alpha_{\text{Fe}} = \alpha_{\text{RE}}$)	0.02	Refs. [1, 46]
Gyromagnetic ratio ($\gamma_{\text{Fe}} = \gamma_{\text{RE}}$)	$1.76 \times 10^{11} \text{ s}^{-1} \text{ T}^{-1}$	Ref. [1]
External magnetic field ($ \mathbf{B}_{\text{ext}} $)	0.12 T	—
Magnetization, $ \mathbf{M}_{\text{Fe}} $	$140 \times 10^3 \text{ J/Tm}^3$	Ref. [1]
Magnetization, $ \mathbf{M}_{\text{RE}} $	$50 \times 10^3 \text{ J/Tm}^3$	Ref. [1]
a_{Fe}^3	$1.2 \times 10^{-28} \text{ m}^3$	Ref. [20]
a_{RE}^3	$8.5 \times 10^{-29} \text{ m}^3$	—

sublattice exchange interaction acts as the primary restoring force. The coexistence of both modes is clearly captured in the time-domain evolution of the Néel vector and is reflected in the Fourier spectrum as two well-separated peaks at 4.2 GHz and 0.48 THz, consistent with the experimental findings of the preceding sections, see Figure 4.4b. The successful reproduction of both modes within the conventional LLG framework under linear excitation confirms that the free energy of Equation (4.8) and the parameters of Table 4.1 provide an accurate description of the equilibrium spin structure and linear dynamics of GdYb-BIG. This validated model now forms the starting point for investigating the nonlinear THz response of the system, where the conventional Zeeman-torque-only description is found to be inadequate, and the inclusion of the FDT becomes essential.

4.5 Nonlinear THz response of the exchange mode

Having established the exchange resonance mode and validated its linear dynamics within the conventional LLG framework, we now turn to the central experimental question of this thesis: whether the nonlinear THz response of GdYb-BIG can be accounted for by the con-

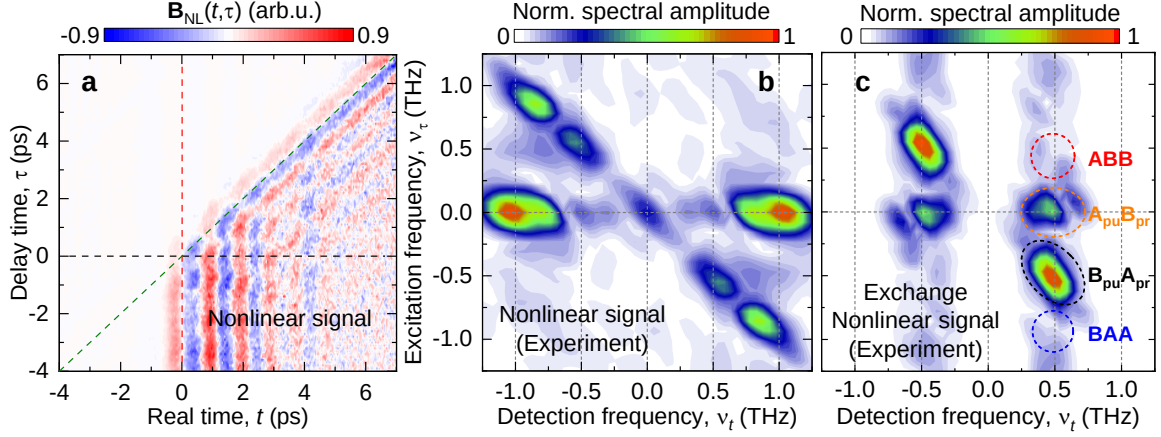


Figure 4.5: **(a)** Contour plot of the total emitted nonlinear THz signal, $B_{NL}(t, \tau)$, as a function of real time and delay time. The green and red dashed lines denote the propagation wavefronts of the two incident THz pulses, while the black dashed line marks the zero-delay position. **(b)** Two-dimensional Fourier spectrum of $B_{NL}(t, \tau)$ plotted as a function of the detection frequency, ν_t , and excitation frequency, ν_τ . The spectrum contains nonlinear contributions from both the Kaplan-Kittel exchange mode at 0.48 THz and the broad material absorption centered around 1 THz. **(c)** Exchange nonlinear spectrum of the Kaplan-Kittel mode obtained after applying a two-dimensional Gaussian filter to the total nonlinear spectrum shown in (b).

ventional Zeeman torque alone, or whether the field-derivative torque is required. Accessing the nonlinear regime requires resonant excitation of the Kaplan-Kittel mode at 0.48 THz with sufficient field amplitude to drive the spin system beyond the linear response. To this end, we employ a dual-pulse 2D-THz spectroscopy scheme, in which two collinear, co-propagating THz pulses A and B, separated by a controlled time delay τ , simultaneously interact with the GdYb-BIG crystal in the presence of an in-plane external field $\mathbf{B}_\perp = 120$ mT applied perpendicular to the THz magnetic field to saturate the sublattice magnetizations.

4.5.1 Extraction of the Nonlinear Signal

When both pulses interact with the sample, the total transmitted field $\mathbf{B}_{AB}(t, \tau)$ contains contributions from each pulse individually as well as from their nonlinear cross-interaction. The purely nonlinear contribution is isolated by subtracting the individual single-pulse re-

sponses from the total two-pulse response,

$$\mathbf{B}_{\text{NL}}(t, \tau) = \mathbf{B}_{\text{AB}}(t, \tau) - \mathbf{B}_{\text{A}}(t, \tau) - \mathbf{B}_{\text{B}}(t), \quad (4.9)$$

where $\mathbf{B}_{\text{A}}(t, \tau)$ and $\mathbf{B}_{\text{B}}(t)$ are the transmitted fields measured with only pulse A and pulse B present, respectively. The individual pulse amplitudes satisfy $|\mathbf{B}_{\text{B}}| \approx 2|\mathbf{B}_{\text{A}}|$, with $|\mathbf{B}_{\text{A}}| \approx 7$ mT and $|\mathbf{B}_{\text{B}}| \approx 14$ mT in our experiments. Since the two pulses interact with the sample collinearly, all nonlinear signals are generated simultaneously and are accessible within the same 2D frequency map. The resulting nonlinear time-domain signal $\mathbf{B}_{\text{NL}}(t, \tau)$ is shown in Figure 4.5a. A two-dimensional Fourier transform with respect to both the real time t and the inter-pulse delay τ then yields the nonlinear spectrum $\mathbf{B}_{\text{NL}}(\nu_t, \nu_\tau)$, where ν_t is the detection frequency at which the nonlinear signal is emitted, and ν_τ is the excitation frequency encoding the delay-time dependence of the nonlinear response, see Figure 4.5b.

4.5.2 Separation of magnetic and non-magnetic contributions

The resulting 2D spectrum, shown in Figure 4.5b, contains two distinct contributions. The first is the nonlinear response of the Kaplan-Kittel exchange mode, centered at 0.48 THz; the second is a broad, non-resonant background arising from the intrinsic material absorption of GdYb-BIG, which peaks near 1 THz and is inherently included in the transmitted THz signal regardless of the magnetic state of the sample. This absorption background, already noted in the linear THz-TDS measurements of the preceding section, complicates direct interpretation of the nonlinear spectrum since the transmitted THz field probes both the resonant magnetic response and the non-resonant dielectric response of the material simultaneously - unlike the magneto-optical Faraday measurement, which is selectively sensitive to the magnetic contribution alone.

To isolate the two contributions, we perform the same dual-pulse measurement in the complete absence of an external magnetic field. Under these conditions, the exchange res-

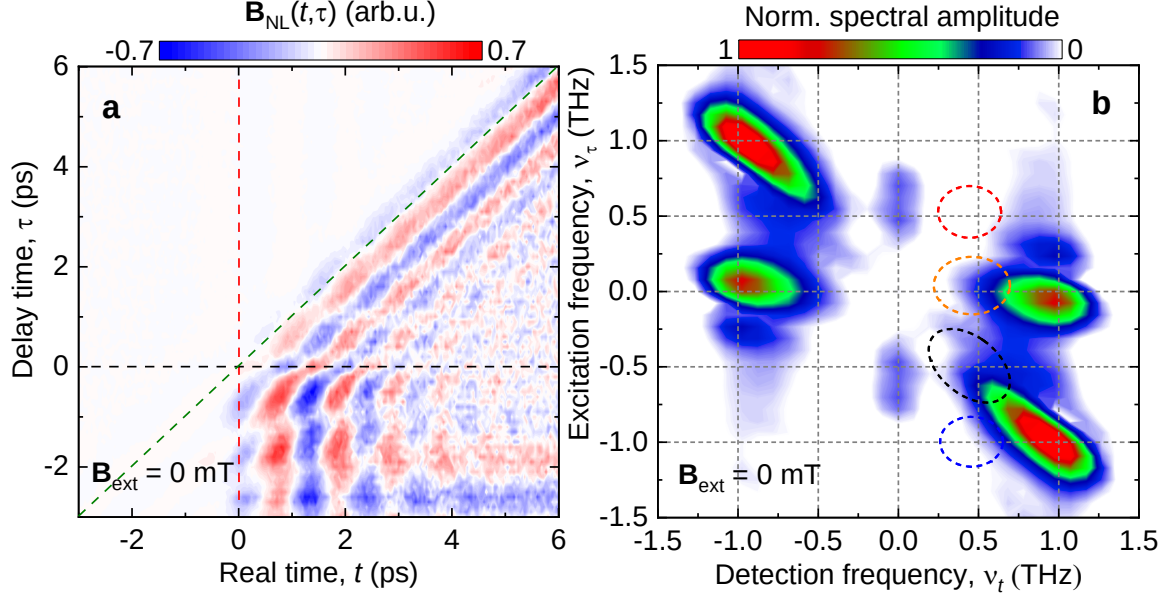


Figure 4.6: **(a)** Contour plot of the nonlinear THz signal emitted from GdYb-BIG in the absence of an external static magnetic field. **(b)** Corresponding two-dimensional Fourier spectrum plotted as a function of the detection frequency, ν_t , and the excitation frequency, ν_τ . The spectrum is dominated by the non-magnetic material response, confirming the suppression of the Kaplan-Kittel exchange mode in the absence of the external magnetic field.

onance is suppressed, and the nonlinear response arises exclusively from the non-magnetic material absorption, as confirmed by the featureless 2D spectrum shown in Figure 4.6a and b. A comparison between the field-on and field-off spectra allows unambiguous identification of the magnetically-driven contribution. A two-dimensional Gaussian spectral filter is then applied to the field-on spectrum to isolate the exchange mode contribution centered at 0.48 THz, removing the absorption background while preserving the spectral lineshape and relative amplitudes of the nonlinear magnetic signals, see Figure 4.5c.

4.5.3 Nonlinear signals and their interpretation

The resulting filtered 2D spectrum, shown in Figure 4.5c, reveals four distinct nonlinear features, all centered at the Kaplan-Kittel resonance frequency $\nu_0 = 0.48$ THz. These signals

arise from third-order, $\chi^{(3)}$, interactions between the incident THz fields and the spin system, in which three field interaction events, involving field components from both pulses, combine to generate the observed nonlinear response. The positions of the four signals in the 2D frequency map can be expressed as linear combinations of the frequency vectors ν_A and ν_B of the two driving pulses.

The two most intense signals are pump-probe signals, in which two interactions with one pulse create a magnon population that is subsequently interrogated by a single interaction with the second pulse. The phase evolution of the pump pulse cancels in this pathway, so that only the phase of the probe pulse contributes to the detected signal. Specifically, the $A_{pu}\text{-}B_{pr}$ signal located at $(\nu_0, 0)$ arises from the frequency combination $\nu_{AB} = \nu_A - \nu_A + \nu_B$, while the $B_{pu}\text{-}A_{pr}$ signal located at $(\nu_0, -\nu_0)$ arises from $\nu_{BA} = \nu_B - \nu_B + \nu_A$.

The two weaker signals are photon-echo-type signals, in which the phase information from both driving pulses is preserved throughout the interaction sequence and partially rephases at the time of detection, analogous to spin echoes in magnetic resonance and photon echoes in nonlinear optics. The ABB-echo signal, located at $(\nu_0, +\nu_0)$, corresponds to the frequency combination $\nu_{ABB} = 2\nu_B - \nu_A$, while the BAA-echo signal, located at $(\nu_0, -2\nu_0)$, corresponds to $\nu_{BAA} = 2\nu_A - \nu_B$. Unlike the pump-probe signals, the echo signals carry direct information about the decoherence timescale of the Kaplan-Kittel resonance mode. This explains the duration over which the collective spin precession maintains phase coherence before dephasing and can therefore be used to extract the magnon decoherence time from the decay of the nonlinear signal along the delay axis τ .

The observation of all four $\chi^{(3)}$ signals in the 2D spectrum constitutes the primary experimental result of this chapter. However, as demonstrated in the following section, these signals cannot be reproduced by the conventional Zeeman-torque-only LLG model: the simulated 2D spectrum in the absence of FDT is essentially featureless, and only upon incorporating the field-derivative torque into the effective field of each sublattice yield a

nonlinear spectrum in quantitative agreement with the experiment.

4.6 Theoretical modeling of the nonlinear spin dynamics: Role of FDT

The experimental 2D THz spectrum presented in the preceding section reveals four distinct $\chi^{(3)}$ nonlinear signals centered at the Kaplan-Kittel resonance frequency of 0.48 THz. A natural first step is to attempt to reproduce these signals within the conventional LLG framework, using the same free energy density and material parameters that successfully described the linear dynamics. Note that, this time, along with the static external field, the Zeeman torque by the THz magnetic field has been added. However, numerical integration of the coupled LLG equations under dual-pulse THz excitation, with the effective field driven solely by the conventional Zeeman torque, yields a 2D spectrum that is essentially featureless: the four nonlinear signals are absent, and no structured spectral weight appears at the exchange resonance frequency, see Figure 4.7c. This failure of the conventional description is not a consequence of an incorrect choice of material parameters - the linear impulsive dynamics are well reproduced by the same model- but rather reflects a fundamental limitation of the non-relativistic LLG equation when applied to systems with high resonance frequencies and appreciable Gilbert damping under strong, rapidly varying THz excitation. In such conditions, relativistic corrections to the spin dynamics become significant, and the conventional Zeeman-torque-only description must be extended.

4.6.1 Incorporating the field-derivative torque

As established in Chapter 2, the relevant relativistic correction in this regime is the field-derivative torque, which arises from the coupling of the spin system to the time derivative of the applied THz magnetic field through the spin-orbit interaction. Within the LLG framework, the FDT enters as a modification of the effective magnetic field of each sublattice

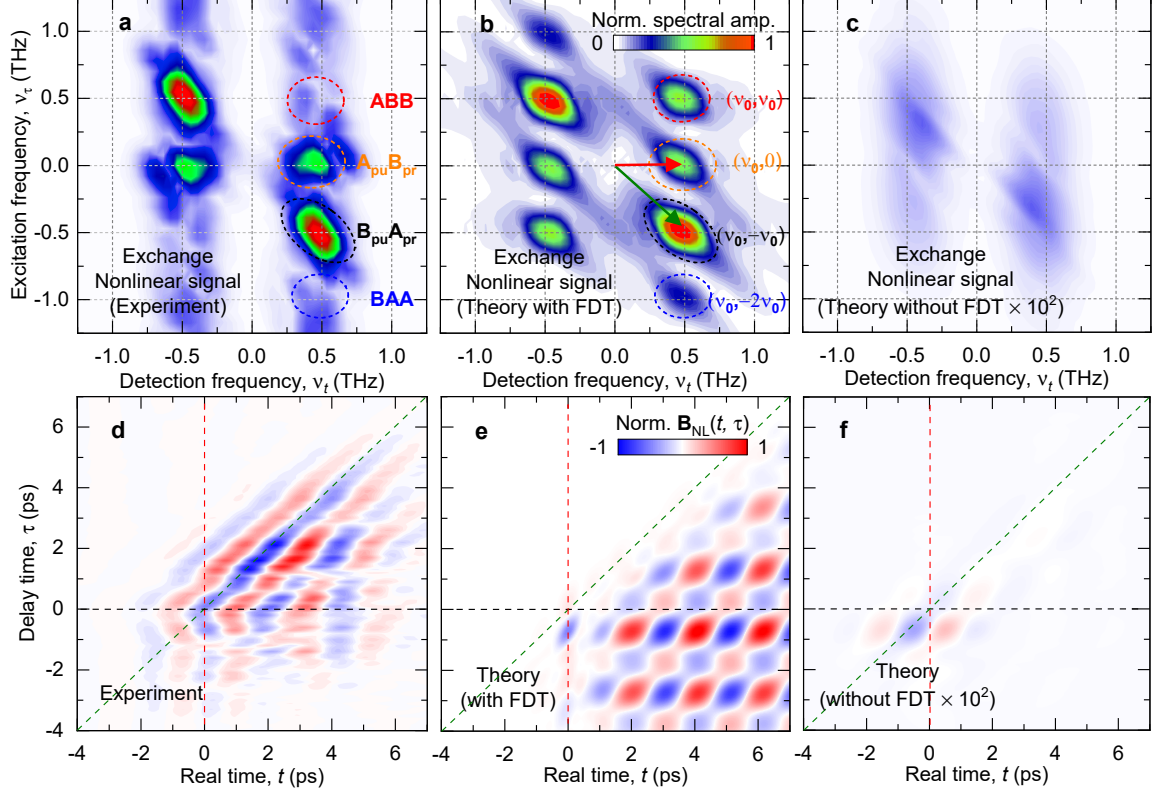


Figure 4.7: **(a)** Normalized two-dimensional exchange nonlinear spectrum of the Kaplan-Kittel mode obtained experimentally. **(b)** Theoretical exchange nonlinear spectrum calculated by incorporating the field-derivative torque (FDT) into the magnetization dynamics, and **(c)** the corresponding spectrum obtained using the conventional LLG equation without FDT. The colored ellipses highlight the positions of the individual nonlinear signals in the 2D frequency spectra. The green and red arrows denote the frequency vectors associated with the incident THz pulses A and B, respectively, while ν_0 represents the Kaplan-Kittel exchange resonance frequency at 0.48 THz. **(d)** Experimental time-domain nonlinear signal obtained by inverse Fourier transformation of the filtered exchange spectrum. **(e)** and **(f)** Corresponding theoretical time-domain nonlinear signals calculated with and without the inclusion of FDT, respectively, plotted as a function of real time (t) and delay time (τ). The green and red dashed lines indicate the propagation wavefronts of THz pulses A and B, respectively, whereas the black dashed line marks the zero-delay position. The spectra shown in **(c)** and the time-domain signal in **(f)** are normalized with respect to **(b)** and **(e)**, respectively.

i,

$$\mathbf{B}_i^{\text{eff}} \longrightarrow \left(\mathbf{B}_i^{\text{eff}} - \frac{\alpha_i a_i^3}{\gamma_i \mu_B \mu_0} \frac{d\mathbf{B}_{\text{THz}}}{dt} \right), \quad (4.10)$$

where a_i^3 is the unit-cell volume per spin for sublattice i , μ_B is the Bohr magneton, μ_0 is the permeability of free space, and $\mathbf{B}_{\text{THz}}$ is the time-dependent THz magnetic field. The structure of the correction in Equation (4.10) encodes the essential material dependence of the FDT: it scales linearly with the Gilbert damping parameter α_i , depends on the unit-cell volume per spin a_i^3 of each sublattice, and is inversely proportional to the gyromagnetic ratio γ_i . Physically, the FDT acts in two complementary ways: it enhances the overall amplitude of the effective driving field relative to the Zeeman-only case, and it introduces an additional frequency-dependent phase into the driving field, shifting the phase relationship between the sublattice responses. Both effects prove essential for generating the $\chi^{(3)}$ nonlinear signals observed experimentally.

4.6.2 Simulated nonlinear spectrum with and without FDT

The modified LLG equations, incorporating the FDT-corrected effective field of Equation (4.10), are solved numerically for both single-pulse and dual-pulse THz excitation schemes. In the single-pulse case, the inclusion of FDT leads to a measurable enhancement of the Kaplan-Kittel mode amplitude in the Néel vector oscillations relative to the conventional Zeeman-torque response, consistent with the theoretical predictions discussed in Chapter 2. In the dual-pulse nonlinear case, the effect of FDT is far more striking: the simulated 2D spectrum now reproduces all four experimentally observed $\chi^{(3)}$ nonlinear signals – the two pump-probe signals and the two echo signals at their correct positions in the 2D frequency map and with their correct relative amplitudes, as shown in Figure 4.7b. In the absence of FDT, the same simulation yields a featureless 2D spectrum, shown in Figure 4.7c, confirming that the nonlinear signals are a direct experimental signature of the relativistic field-derivative torque rather than of higher-order Zeeman nonlinearities. A sim-

ilar conclusion can be drawn from the time-domain nonlinear response. When the FDT is incorporated into the simulations, the calculated exchange nonlinear signal reproduces the experimental observations remarkably well, accurately capturing both the oscillation period of the Kaplan-Kittel mode and its nonlinear evolution along the t and τ axes. In contrast, simulations based solely on the conventional LLG equation fail to resolve the nonlinear exchange dynamics in the time domain, as illustrated in Figures 4.7d-f.

The origin of this qualitative difference can be traced to the role of the FDT in satisfying the conditions required for third-order nonlinear signal generation. The FDT introduces a frequency-dependent phase shift into the effective driving field, which modifies the phase-matching conditions governing the nonlinear interaction between the two THz pulses and the exchange magnon. In addition, the overall amplitude of the frequency-dependent driving field is enhanced by the FDT contribution, ensuring that the threshold field amplitude required for generating distinct $\chi^{(3)}$ signals is met. Both conditions are provided exclusively by the FDT term in the present system; without it, neither the phase-matching nor the amplitude threshold conditions are satisfied, and no structured nonlinear spectrum emerges.

4.6.3 Relative magnitude of FDT and material dependence

A quantitative comparison of the FDT and conventional Zeeman torque contributions can be obtained from the FDT correction to the effective magnetic field in Eq. The conventional Zeeman contribution to the effective driving field is proportional to the input THz magnetic field, whereas the FDT contribution is proportional to its time derivative. From there we can see, the relative magnitude of the FDT contribution increases linearly with both the Gilbert damping parameter and the excitation frequency. Using the material parameters employed in the simulations, the material-dependent coefficient entering the FDT contribution is of the order of 10^{-11} s. Consequently, the relative magnitude can be expressed in the reduced form

$$\left| \frac{\text{FDT}}{\text{ZT}} \right| \approx 10^{-11} \text{s} \cdot \alpha \cdot \nu_0, \quad (4.11)$$

where ν_0 is the exchange resonance frequency. For GdYb-BIG, with $\alpha = 0.02$ and $\nu_0 = 0.48$ THz, this ratio evaluates to approximately 0.1, indicating that the FDT modifies the effective driving field by approximately 10% relative to the Zeeman torque. Although this relative correction is modest in magnitude, it can have a substantial effect on the nonlinear response because the FDT modifies both the amplitude and phase of the effective THz driving field. As demonstrated in Section 4.6.2, this modification is sufficient to produce the structured $\chi^{(3)}$ nonlinear response observed experimentally, whereas the corresponding Zeeman-torque-only calculation does not reproduce these features.

The strength of the FDT contribution is strongly material-dependent. From Equation (4.11), it is clear that larger damping parameters and higher exchange resonance frequencies both amplify the FDT contribution relative to the Zeeman torque. Equally important is the equivalence of the sublattice gyromagnetic ratios. In GdYb-BIG, $\gamma_{\text{Fe}} = \gamma_{\text{RE}}$, which maximizes the relative FDT contribution by preventing the conventional Zeeman torque from dominating one sublattice's response over the other. In systems where the sublattice gyromagnetic ratios differ appreciably, such as in the ferrimagnetic garnet $\text{Tm}_2\text{BiFe}_{4.2}\text{Ga}_{0.8}\text{O}_{12}$ [20], where inequivalent gyromagnetic ratios cause the Zeeman torque to dominate, the FDT contribution is suppressed and the nonlinear signals do not exhibit the characteristic pattern observed here. These findings establish a clear set of material design criteria – high damping, THz-frequency exchange resonance, and equivalent sublattice gyromagnetic ratios, for identifying and engineering future magnetic systems in which relativistic spin-torque effects can be experimentally accessed and exploited. A systematic theoretical exploration of how these material parameters individually govern the FDT-induced nonlinear response is presented in the next chapter.

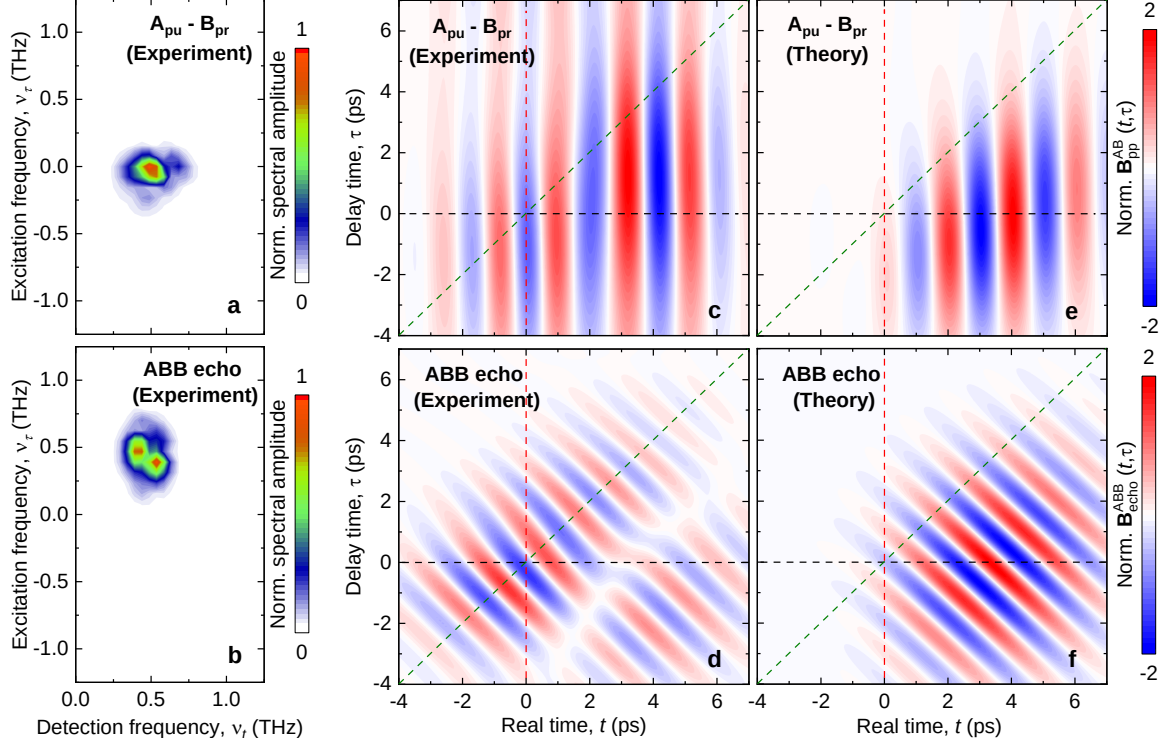


Figure 4.8: **(a)** Two-dimensional filtered spectrum corresponding to the A_{pu} - B_{pr} pump-probe signal and **(b)** the ABB photon-echo signal. **(c)** and **(d)** Time-domain contour plots of the inverse Fourier-transformed nonlinear signals, $B_{pp}^{AB}(t, \tau)$ and $B_{echo}^{ABB}(t, \tau)$, respectively. **(e)** and **(f)** Corresponding theoretical time-domain contour plots obtained from the numerical simulations. The green and red dashed lines denote the propagation wavefronts of the incident THz pulses A and B, respectively, while the black dashed line marks the zero-delay position.

4.7 Signatures of FDT in the nonlinear exchange dynamics

The clear spatial separation of the four $\chi^{(3)}$ nonlinear signals in the 2D frequency map, achieved through the Gaussian filtering procedure described in the preceding section, enables a detailed analysis of their individual temporal characteristics free from interference by the non-magnetic absorption background. To this end, we selectively isolate the A_{pu} - B_{pr} pump-probe signal and the ABB echo signal by applying individual 2D Gaussian spectral filters centered at their respective positions in the 2D frequency map, and transform each filtered component back into the time domain through an inverse Fourier transform. The re-

sulting time-resolved signals are then compared directly against the theoretical predictions of the FDT-modified LLG model.

4.7.1 Time-domain analysis and comparison with theory

The experimentally extracted time-domain traces of the A_{pu} - B_{pr} and ABB echo signals are shown in Figures 4.8c and 4.8d, alongside the corresponding theoretical signals from the FDT-modified LLG model in Figures 4.8e and 4.8f. A close qualitative agreement is observed between experiment and theory, particularly in the evolution of the pulse fronts in the (t, τ) plane, which are the most sensitive indicator of the phase relationship between the two driving pulses and the nonlinear spin response. This agreement is obtained exclusively when FDT is incorporated into the model; the conventional Zeeman-torque-only simulation fails to reproduce the temporal structure of either signal.

A notable feature of the B_{pu} - A_{pr} signal, see Figure 4.7a is its comparatively broader spectral profile relative to the other three nonlinear signals, indicative of inhomogeneous broadening. This asymmetric linewidth arises from the unequal amplitudes of the two driving pulses ($|B_B| \approx 2|B_A|$), which produces a spatial inhomogeneity in the effective magnetic field experienced by the two sublattices across the sample, giving rise to a distribution of local resonance conditions. This inhomogeneous contribution is, however, comparatively weak relative to the dominant homogeneous broadening of the exchange resonance and does not affect the interpretation of the other nonlinear signals, see Section 4 of Appendix A.

4.7.2 Magnon population decay and decoherence times

Beyond the qualitative comparison of pulse-front dynamics, the nonlinear signals encode quantitative information about the collective spin dynamics of GdYb-BIG that is inaccessible from the linear measurements alone. In particular, the real-time oscillation of the nonlinear magnetization is directly traced along the t -axis of the 2D map, while the coher-

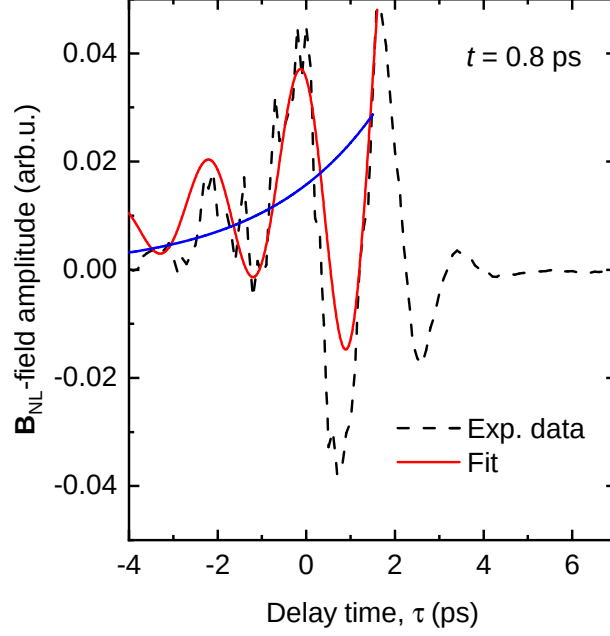


Figure 4.9: The nonlinear $\mathbf{B}_{\text{NL}}$ FID signal as a function of delay time taken at a fixed real time of $t = 0.8$ ps (black dashed line). The entire fit is shown by the red solid line, with the decay of the carrier population shown by the blue solid line.

ence of the spin system manifests as a short-lived oscillatory feature along the τ -axis. A line scan of $\mathbf{B}_{\text{NL}}(t, \tau)$ along the τ -axis at a fixed real time $t = 0.8$ ps provides a direct experimental trace of the delay-time dependence of the nonlinear signal, from which both the magnon population decay time T_1 and the magnon decoherence time T_2^* can be extracted simultaneously, see Figure 4.9.

To extract these timescales quantitatively, the τ -dependent nonlinear trace is modeled using the function [105]

$$\mathbf{B}_{\text{NL}}(\tau) = A_1 \exp\left(-\frac{\tau}{T_1}\right) + A_2 \sin(\omega\tau) \exp\left(-\frac{\tau}{T_2^*}\right), \quad (4.12)$$

where the first term represents the exponential decay of the magnon population with characteristic time T_1 , and the second term represents the oscillatory, exponentially decaying nonlinear magnetization associated with the coherent magnon precession, characterized

by the decoherence time T_2^* . Physically, T_1 corresponds to the duration over which the magnon population created by the pump pulse persists before relaxing back to equilibrium – the time window within which the probe pulse can still interact with a non-equilibrium magnon population to generate a nonlinear response. The decoherence time T_2^* , by contrast, characterizes the duration over which the collective spin precession maintains phase coherence under the two-pulse excitation before the individual spin contributions dephase and the macroscopic nonlinear signal decays. It is the period within which THz-probe-induced spin rephasing can occur, and the echo-type signals can be observed.

Fitting Equation (4.12) to the experimental τ -dependent trace yields a magnon population decay time of $T_1 = 6.1 \pm 2.4$ ps and a magnon decoherence time of $T_2^* = 2.5 \pm 0.5$ ps. Both timescales lie firmly in the picosecond range, demonstrating that GdYb-BIG supports coherent exchange spin dynamics over durations far exceeding the THz pulse duration itself. The fact that $T_1 > T_2^*$, consistent with the general relation $T_2^* \leq 2T_1$, confirms that dephasing, rather than population decay, is the dominant mechanism limiting the coherence of the Kaplan-Kittel resonance in this system.

4.7.3 Material specificity of FDT and comparison with related systems

The pronounced manifestation of FDT in GdYb-BIG raises the question of why similar effects were not reported in previous nonlinear THz spectroscopy studies on related magnetic systems, including rare-earth orthoferrites such as YFeO_3 and iron garnets such as $\text{Tm}_2\text{BiFe}_{4.2}\text{Ga}_{0.8}\text{O}_{12}$. The answer lies in the material-specific conditions required for FDT to become observationally significant, as established in the preceding theoretical modeling section.

In YFeO_3 , the Gilbert damping parameters are $\alpha = 1.3 \times 10^{-3}$ for the antiferromagnetic mode and $\alpha = 5.5 \times 10^{-4}$ for the ferromagnetic mode – one to two orders of magnitude smaller than the value of $\alpha = 0.02$ in GdYb-BIG. Since the FDT-to-Zeeman-torque ratio

scales as $|\text{FDT}/\text{ZT}| \propto \alpha \cdot \nu_0$, such low damping values render the FDT contribution negligibly small, and it is entirely inconsequential to invoke it in explaining the nonlinear THz spectra of YFeO_3 [36, 37]. This is confirmed numerically: simulations of the antiferromagnetic spin dynamics of YFeO_3 under single-pulse THz excitation, shown in Figure 4.10a, demonstrate that the inclusion or exclusion of FDT makes virtually no difference to the dynamics. By sharp contrast, the equivalent simulation for GdYb-BIG , shown in Figure 4.10b, reveals a dramatic enhancement of the Néel-vector oscillation amplitude.

To further substantiate that the FDT criterion is not specific to GdYb-BIG but is a general material condition, we hypothetically increase the damping parameter of YFeO_3 by an order of magnitude to bring it into the regime required for FDT. As shown in Figure 4.10c, at such elevated damping, the antiferromagnetic dynamics of YFeO_3 are strongly suppressed, and the oscillations become overdamped. The FDT correction does lead to a measurable increase in the oscillation amplitude and an additional phase shift under these hypothetical conditions, which is consistent with the general predictions of the relativistic LLG framework. This result highlights a fundamental material constraint: the FDT is most observable in a system that simultaneously maintains a well-defined, high-frequency exchange magnon mode and possesses a damping parameter large enough for the FDT contribution to be significant - a combination that, among known magnetic systems, GdYb-BIG uniquely satisfies at room temperature. The systematic exploration of how specific material parameters - damping and sublattice spin volume individually govern the FDT-induced nonlinear response forms the subject of the following chapter.

4.8 Other possible sources of nonlinearity

The observation of the nonlinear $\chi^{(3)}$ response is not, by itself, uniquely indicative of the field-derivative torque, since other nonlinear mechanisms may contribute to the ob-

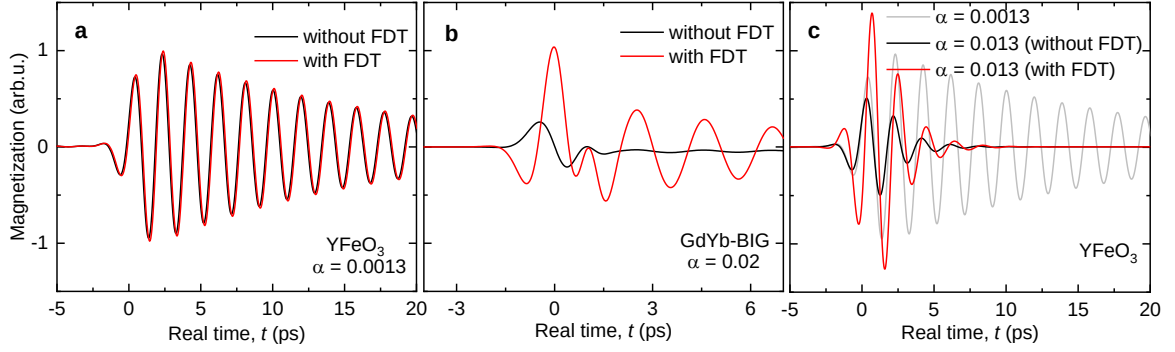


Figure 4.10: **(a)** Simulated antiferromagnetic magnetization dynamics of YFeO_3 calculated with and without the inclusion of the field-derivative torque (FDT). **(b)** Corresponding magnetization dynamics of GdYb-BIG under identical conditions, highlighting the pronounced influence of FDT. **(c)** Comparison of the simulated magnetization dynamics in YFeO_3 obtained by hypothetically increasing the Gilbert damping by one order of magnitude and incorporating the FDT.

served response. In particular, transient heating and the associated modification of magnetic anisotropy or exchange parameters, as well as nonlinear spin-wave scattering processes such as Suhl instabilities, can, in principle, generate nonlinear magnetic signals. First, regarding the transient thermal effects, in the present experiment, the exchange mode is driven resonantly and directly by the magnetic-field component of the THz pulse through the Zeeman interaction. In contrast to optical excitation, where absorption of an ultrafast optical pulse can deposit substantial energy into the electronic and lattice systems and thereby induce transient changes in magnetic parameters, the THz excitation used here primarily couples directly to the magnetic degrees of freedom. The relatively low THz-field energy deposited in the sample does not provide a significant thermal perturbation to the system. Therefore, transient heating is not expected to be the dominant source of the observed nonlinear response. Furthermore, in the nonlinear LLG simulations, the experimentally determined room-temperature parameters, including the anisotropy energy and magnetic moments, are kept fixed, so that the calculated nonlinear response does not arise from a pump-induced thermal modification of these parameters.

Second, we considered the possibility of nonlinear spin-wave scattering or a Suhl-type instability. A Suhl instability is a nonlinear parametric process in which a strongly driven, long-wavelength magnetic mode becomes unstable and transfers energy to other spin-wave modes, typically involving finite-wave-vector excitations. In the context of the present 2D-THz experiment, such a process would be expected to redistribute part of the energy of the coherently driven 0.48 THz Kaplan-Kittel exchange mode into additional magnon modes. This could give rise to additional spectral features at shifted detection frequencies in the two-dimensional spectrum.

In the present experiment, however, the nonlinear response remains concentrated around the 0.48 THz, which is the exchange (Kaplan-Kittel) mode and consists of four well-defined features corresponding to the pump-probe and photon-echo signals. These features arise from the nonlinear interaction of the two THz pulses with the coherently driven exchange mode. We do not observe additional spectral branches or distinct shifted-frequency features that would indicate a significant transfer of energy from the 0.48 THz mode to other magnon modes within the measured frequency range. Thus, while such nonlinear processes cannot be completely excluded, the experimental data do not indicate that they are the dominant origin of the observed four $\chi^{(3)}$ features.

4.9 Summary

In this chapter, we have presented the first experimental demonstration of the relativistic field-derivative torque in the nonlinear THz magnetization dynamics of the ferrimagnetic iron garnet GdYb-BIG. Starting from the material characterization and the theoretical framework of the two-sublattice LLG equation, we identified the Kaplan-Kittel exchange resonance mode at 0.48 THz through complementary linear THz time-domain spectroscopy and THz-pump optical-probe Faraday rotation measurements. The conventional LLG

framework, employing the free energy density incorporating exchange, anisotropy, Zeeman, and demagnetization contributions, was shown to successfully reproduce both the low-frequency BVMW mode at 4.2 GHz and the high-frequency Kaplan-Kittel mode at 0.48 THz under linear impulsive excitation, validating the material parameters used throughout this chapter. Turning to the nonlinear regime, resonant dual-pulse 2D-THz spectroscopy of the Kaplan-Kittel mode revealed four distinct $\chi^{(3)}$ nonlinear signals - two pump-probe and two photon-echo signals in the two-dimensional frequency map. Crucially, these signals could not be reproduced by the conventional Zeeman-torque-only LLG equation under any choice of physically reasonable parameters, while their positions, relative amplitudes, and time-domain pulse-front structure were qualitatively reproduced upon incorporating the field-derivative torque into the effective field of each sublattice. A quantitative estimate of the FDT-to-Zeeman-torque ratio yields a value of approximately 0.1 for GdYb-BIG, sufficient to generate qualitatively distinct spectral features that are entirely absent in the conventional description. Analysis of the free-induction decay of the nonlinear signal along the delay axis further allowed the extraction of the magnon population decay time ($T_1 = 6.1 \pm 2.4$ ps) and the magnon decoherence time ($T_2^* = 2.5 \pm 0.5$ ps), both in the picosecond range, confirming that GdYb-BIG supports coherent exchange spin dynamics over timescales sufficient for THz spintronic applications. Finally, a comparative analysis with related magnetic systems, including the antiferromagnet YFeO₃ demonstrated that the observability of FDT is strongly material-dependent, requiring the simultaneous satisfaction of three conditions: a THz-frequency exchange resonance, a Gilbert damping parameter $\alpha \gtrsim 0.01$, and equivalent sublattice gyromagnetic ratios. GdYb-BIG uniquely satisfies all three at room temperature, making it the ideal platform for this investigation. The systematic theoretical exploration of how these material parameters individually and collectively govern the FDT-induced nonlinear response, in particular the role of sublattice spin volume ratio and damping threshold, forms the subject of the

following chapter.

Chapter 5

Role of material-dependent properties in THz field-derivative-torque-induced nonlinear magnetization dynamics

The experimental results of the preceding chapter established that the relativistic field-derivative torque is indispensable for describing the nonlinear THz magnetization dynamics of GdYb-BIG, and that its observability is governed by a specific combination of material conditions - a THz-frequency exchange resonance, a comparatively large Gilbert damping parameter, and equivalent sublattice gyromagnetic ratios. While the experimental demonstration confirms the existence of FDT as a physically relevant torque in a real ferrimagnetic system, it also raises a question: how does the FDT contribution evolve as the intrinsic material parameters (such as: sublattice spin-volume ratio and Gilbert damping) are systematically varied?

Addressing this question requires a systematic theoretical investigation that goes beyond the fixed material parameters of GdYb-BIG. The present chapter provides this investigation through comprehensive numerical modeling of both linear and nonlinear THz magnetization dynamics within the FDT-modified LLG framework developed in Chapter 4. By independently varying the sublattice spin-volume ratio and Gilbert damping, we identify the precise material conditions that govern the strength and observability of the relativistic FDT contribution. These results not only deepen the physical understanding of the experimental findings of Chapter 4, but also establish concrete design criteria for identifying and engineering future ferrimagnetic materials in which relativistic spin-torque effects can be accessed and exploited at THz frequencies.

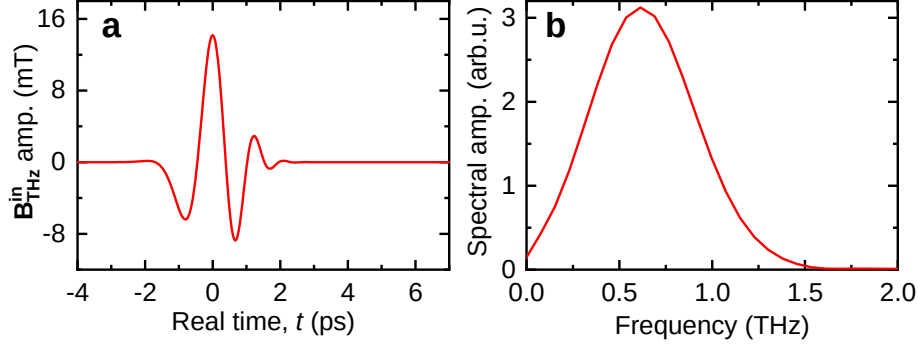


Figure 5.1: **(a,b)** Modeled THz pulse and its corresponding frequency spectrum, respectively.

5.1 Sublattice dynamics

To model the THz-driven magnetization dynamics, we consider a single-cycle THz magnetic field pulse applied along the x -direction, with temporal profile governed by [44, 48]

$$\mathbf{H}_{\text{THz}}(t) = H_0 \cos(2\pi f_0 \Gamma [e^{t/\Gamma} - 1]) e^{-t^2/\sigma^2} \hat{\mathbf{x}}, \quad (5.1)$$

where H_0 is the pulse amplitude, $f_0 = 0.6$ THz is the center frequency, $\sigma = 1$ ps is the pulse duration, and $\Gamma = 2.6$ ps is the chirp parameter, see Figure 5.1a,b. These parameters are chosen to closely reproduce the experimental THz pulse conditions of Chapter 4, ensuring direct comparability between the theoretical modeling and experimental observations. A static external magnetic field, $\mathbf{B}_{\text{ext}} = 120$ mT is applied along the y -direction, perpendicular to the THz field, to saturate the sublattice magnetizations and establish a well-defined ferrimagnetic ground state prior to each excitation.

5.1.1 Effect of FDT on the effective driving field

The applied THz field drives the magnetization dynamics primarily through the conventional Zeeman torque (ZT). When the field-derivative torque is additionally incorporated, the effective driving field of each sublattice acquires the additional relativistic correction of

Equation (4.10), proportional to $d\mathbf{B}_{\text{THz}}/dt$. The resulting effective fields with and without FDT are shown in Figure 5.2a. Two consequences of the FDT correction are immediately apparent from inspection of the field profiles. First, the overall amplitude of the combined ZT + FDT effective field is substantially enhanced relative to the Zeeman-only field, since the time-derivative contribution adds constructively in the high-frequency components of the THz pulse. Second, the FDT introduces an additional frequency-dependent phase into the effective driving field, visible as a shift in the resonance frequency evident in the Fourier spectra of Figure 5.2b.

5.1.2 Exchange dynamics: Néel vector response

The magnetization dynamics are most conveniently analyzed in terms of the antiferromagnetic Néel vector $\mathbf{L} = \mathbf{M}_{\text{RE}} - \mathbf{M}_{\text{Fe}}$, which serves as the natural order parameter for the anti-phase exchange precession of the two sublattices. Upon single-pulse THz excitation, the system exhibits coherent exchange oscillations at the Kaplan-Kittel resonance frequency, corresponding to the anti-phase precession of the RE and Fe sublattice magnetizations around the effective field. As shown in Figure 5.2c, the inclusion of FDT leads to a substantial enhancement in the amplitude of the Néel vector oscillations relative to the Zeeman-only case, while the overall phase of the time-domain response remains largely unchanged. This seemingly counterintuitive result – the input effective fields differ significantly in phase between the ZT and ZT + FDT cases, yet the dynamical response phases are nearly the same – reflects the fact that the intrinsic precessional dynamics of the ferrimagnetic system are governed by the effective field landscape shaped by both torques together, and that the additional phase introduced by the FDT into the driving field is partially absorbed into the enhanced oscillation amplitude rather than manifesting as a phase shift of the Néel vector.

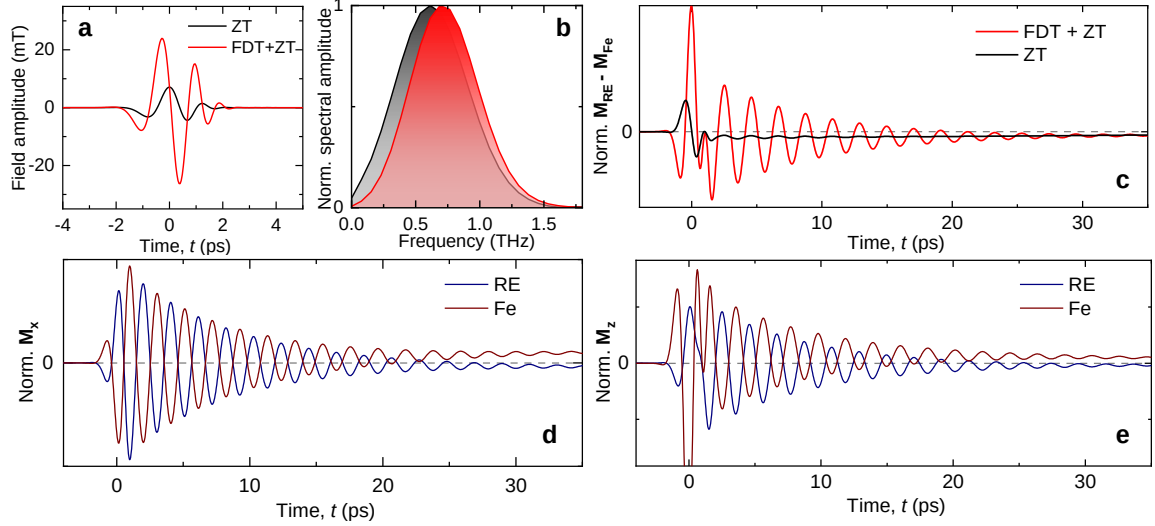


Figure 5.2: **(a)** Modeled effective magnetic field corresponding to the Zeeman torque (ZT) and the total effective field obtained after incorporating the field-derivative torque (FDT). **(b)** Normalized Fourier spectra of the fields shown in (a), illustrating the frequency shift introduced by the inclusion of FDT. **(c)** Simulated exchange dynamics of the rare-earth and Fe sublattice magnetizations calculated with and without the FDT. **(d)** and **(e)** Simulated temporal evolution of the x - and z -components, respectively, of the individual sublattice magnetizations in the presence of FDT.

5.1.3 Sublattice-resolved magnetization

A more detailed picture emerges from examining the individual sublattice magnetizations. As shown in Figures 5.2d and 5.2e, the transverse (x -direction) components of the RE and Fe sublattice magnetizations oscillate with equal amplitude but in anti-phase with each other, as expected for the antiferromagnetically coupled exchange mode in which the two sublattices precess in opposite directions. In contrast, the out-of-plane (z -direction) components of the two sublattice magnetizations exhibit unequal amplitudes, reflecting the difference in the effective fields experienced by the individual sublattices arising from their distinct unit-cell volumes per spin, $a_{\text{Fe}}^3 \neq a_{\text{RE}}^3$, which enter asymmetrically into the FDT correction of Equation (4.10).

At timescales beyond the initial THz excitation window, a small residual magnetization

component persists along the z -direction. This long-lived contribution arises from the low-frequency backward-volume magnetostatic wave (BVMW) mode at 4.2 GHz, which is also driven by the THz pulse through the same Zeeman mechanism but oscillates slowly relative to the picosecond observation window and therefore appears as a quasi-static offset. It is worth noting that the present simulations do not account for the broadband material absorption of GdYb-BIG, which contributes a non-resonant background to the experimental THz signals and may modify the long-time dynamics quantitatively, though not qualitatively. The successful reproduction of the key dynamical features, the exchange oscillation amplitude, the sublattice phase relationship, and the FDT-induced amplitude enhancement, demonstrates that the FDT-modified LLG framework provides a complete and accurate description of the single-pulse linear response of GdYb-BIG, and sets the basis for the systematic material-parameter study presented in the following sections.

5.2 Effect of material properties

5.2.1 Volume per spin

In ferrimagnetic systems, the relative importance of FDT is strongly influenced by the ratio of the unit-cell volume per spin of the two sublattices. To explore this effect, we systematically vary the ratio of the volumes associated with the iron and rare-earth sublattices and analyze the resulting exchange dynamics. We find that the amplitude of the Néel vector oscillations is generally enhanced in the presence of FDT, except when the volume ratio approaches unity. In this symmetric limit, the system exhibits a reduced sensitivity to FDT, accompanied by a noticeable phase shift in the oscillations. This behavior is reminiscent of antiferromagnetic systems, although the phase shift deviates from the ideal value due to the inherent imbalance in sublattice magnetizations.

In the present theoretical analysis, the ratio ($a_{\text{Fe}}^3/a_{\text{RE}}^3$) is varied independently in order

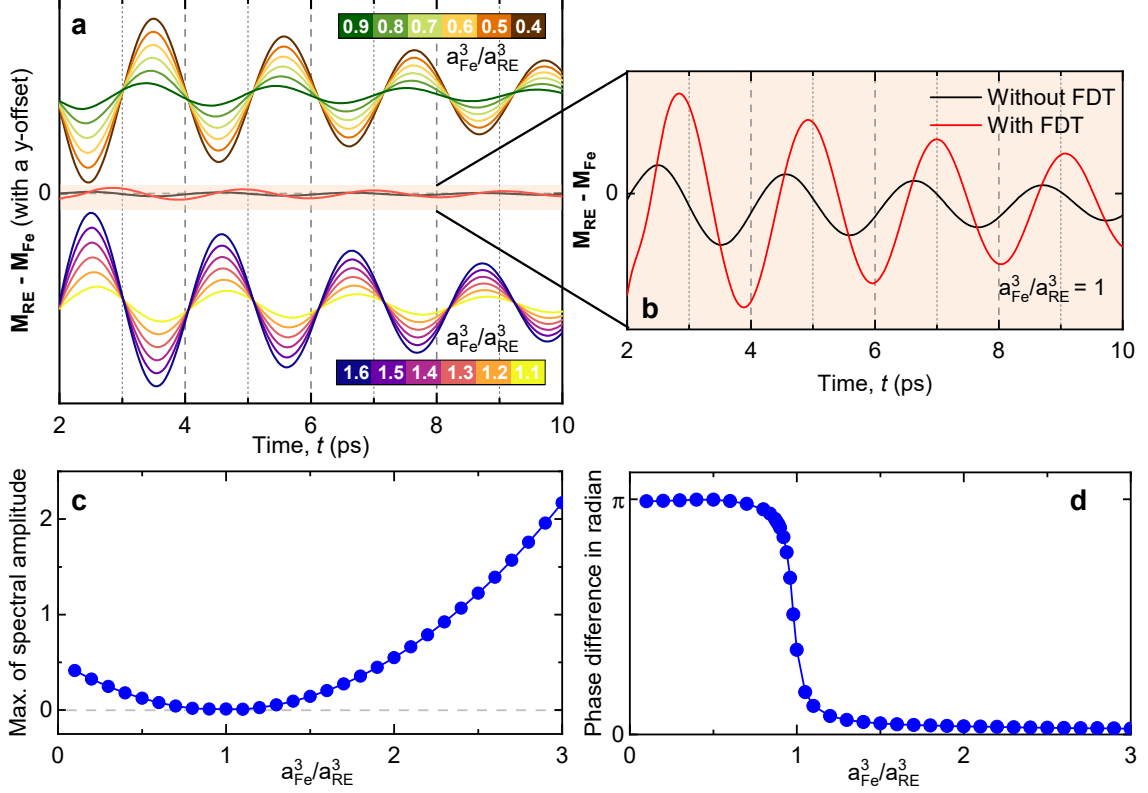


Figure 5.3: **(a)** Simulated THz-induced exchange dynamics of the ferrimagnetic system calculated with and without the inclusion of the field-derivative torque (FDT) for different ratios of the unit-cell volume per spin of the Fe and rare-earth (RE) sublattices. The black curve represents the Néel vector dynamics in the absence of FDT, while the colored curves correspond to the dynamics obtained with FDT for different values of a_{Fe}^3/a_{RE}^3 . **(b)** Magnified view of the Néel vector dynamics for the case $a_{Fe}^3 = a_{RE}^3$, highlighting the difference between the simulations performed with and without FDT. **(c)** Variation of the maximum spectral amplitude, extracted from the Fourier spectra of the time-domain signals shown in (a), as a function of the volume ratio. **(d)** Phase difference between the Néel vector oscillations calculated with and without FDT as a function of the unit-cell volume ratio.

to isolate its influence on the FDT-induced dynamics. In a real material, however, this quantity is determined by the crystal structure, the number of magnetic ions occupying the respective sublattices, and their site occupancy, and therefore cannot generally be varied independently while keeping all other material parameters unchanged. The variation considered here should therefore be regarded as a systematic theoretical study of the role of sublattice asymmetry rather than as a directly tunable experimental parameter. Differ-

ent degrees of sublattice asymmetry can nevertheless be realized across different ferrimagnetic compounds through variations in crystal structure, chemical substitution, rare-earth-ion selection, and magnetic-site occupancy. Thus, the dependence identified here provides a framework for assessing how structural and compositional differences between ferrimagnetic materials may influence the relative importance of FDT.

A broader analysis across different volume ratios reveals a clear dependence of both the amplitude and phase of the exchange oscillations on sublattice asymmetry. When the volume associated with the iron sublattice is smaller than that of the rare-earth sublattice, the system exhibits a phase reversal in the dynamics compared to the case without FDT. Conversely, when the iron sublattice volume is larger, the phase remains largely unchanged, see Figure 5.3a,b. The transition between these regimes occurs near the point where the two volumes become equal, indicating an underlying symmetry associated with the approach to an antiferromagnetic-like configuration 5.3d. Additionally, the amplitude of the exchange oscillations shows a pronounced dip near this symmetric point, further highlighting the reduced effectiveness of FDT in this regime, see Figure 5.3c.

These findings provide important insights into how material-specific parameters can be used to control the phase and amplitude of ultrafast magnetization dynamics. In particular, the sensitivity of the system to the sublattice volume ratio suggests that careful material design can be employed to tailor the influence of relativistic torques. For the investigation of nonlinear dynamics discussed in the subsequent section, we focus on a representative volume ratio greater than unity, where the effects of FDT are pronounced and stable. Notably, beyond a certain threshold value of this ratio, the nonlinear response does not exhibit significant additional changes, indicating a saturation of the FDT-induced effects.

Having established the role of sublattice volume asymmetry in governing the influence of FDT on the exchange dynamics, we now turn our attention to another key material parameter, Gilbert damping.

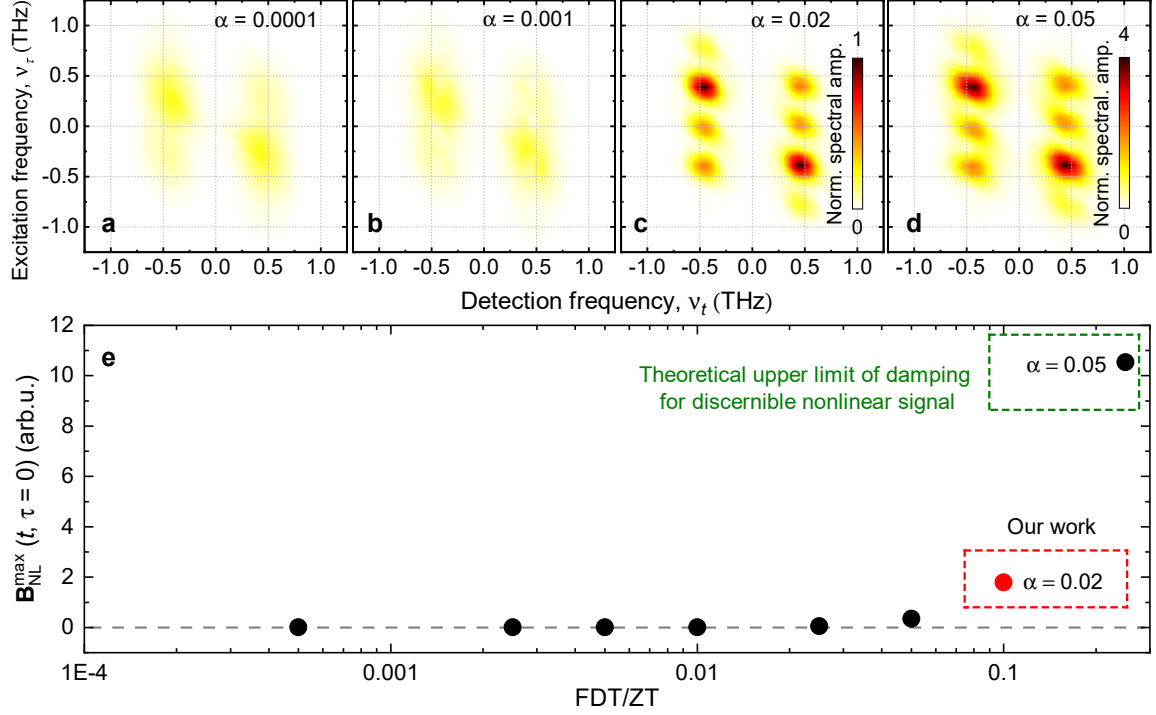


Figure 5.4: **(a–d)** Simulated two-dimensional exchange nonlinear spectra of the Kaplan-Kittel mode calculated for different values of the Gilbert damping parameter. **(e)** Variation of the maximum nonlinear signal amplitude, $B_{NL}^{\max}$, as a function of the Gilbert damping, showing a non-monotonic dependence. The red data point corresponds to the experimental conditions employed in the present study.

5.2.2 Gilbert damping

We now examine the influence of the Gilbert damping parameter on the evolution of the nonlinear magnetization response. In this analysis, the exchange resonance frequency is kept fixed, while the damping parameter is systematically varied for a representative unit-cell volume ratio. Since the relative strength of the FDT with respect to the ZT scales linearly with both the damping parameter α and the resonance frequency, tuning α provides a direct means to probe the impact of relativistic effects on the nonlinear dynamics.

To quantify this behavior, we evaluate the maximum amplitude of the nonlinear signal as a function of the ratio between FDT and ZT. Our results reveal an approximate crossover

in the damping regime beyond which distinct nonlinear features begin to emerge clearly, see Figure 5.4. For very small damping values, typical of antiferromagnetic systems, the contribution of FDT remains negligible. Consequently, the nonlinear response in such systems is predominantly governed by the Zeeman torque, and the inclusion of FDT does not lead to any appreciable modification of the nonlinear spectra.

In contrast, as the damping parameter increases, the relative contribution of FDT becomes more significant, leading to a pronounced enhancement of the nonlinear signal. Interestingly, this enhancement does not follow a simple monotonic trend; instead, the nonlinear response exhibits a non-monotonic dependence on the damping parameter. This indicates a complex interplay between dissipation and coherent spin dynamics in determining the overall signal strength, see Figure 5.4.

The non-monotonic dependence of the nonlinear response on the Gilbert damping can be understood from the competing roles of damping in the FDT contribution and in the coherent exchange dynamics. The magnitude of the FDT relative to the conventional Zeeman torque increases approximately linearly with the damping parameter, $|\text{FDT}/\text{ZT}| \propto \alpha\nu_0$. Thus, increasing α initially enhances the relativistic contribution to the effective driving field, resulting in an increase in the nonlinear response. At the same time, however, increasing damping introduces stronger dissipation into the magnetization dynamics, reducing the lifetime and coherence of the exchange-mode precession. At sufficiently large α , the suppression of coherent exchange dynamics becomes dominant over the enhancement of the FDT contribution, leading to a reduction and eventual distortion of the nonlinear response, as illustrated in Figure 5.5.

To quantify this competition, the temporal weight of the exchange oscillation was evaluated by integrating the oscillatory response over the time window 2.0-6.2 ps. The resulting temporal weight is plotted as a function of the Gilbert damping parameter in Figure 5.6. The temporal weight increases with increasing damping from a nearly negligible value at low

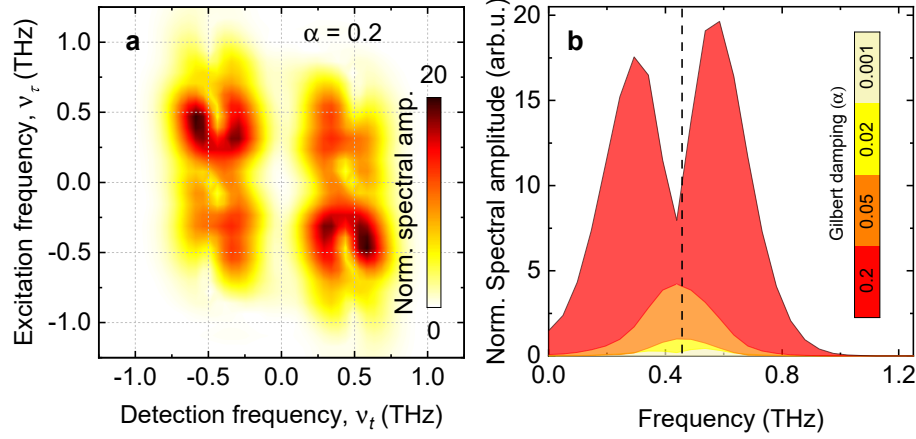


Figure 5.5: **(a)** Simulated two-dimensional exchange nonlinear spectrum of the Kaplan-Kittel mode for a hypothetical Gilbert damping of $\alpha = 0.2$. At such a high damping, the nonlinear spectral features become significantly distorted, making the individual nonlinear signals difficult to distinguish. **(b)** Normalized spectral amplitude of the exchange nonlinear mode for different values of the Gilbert damping parameter. The line profiles are extracted by taking horizontal cuts at $\nu_\tau = -0.4$ THz from the nonlinear spectra shown in (a) and Figures 5.4(b–d).

damping and reaches a maximum around $\alpha = 0.05$. This increase is consistent with the progressive enhancement of the FDT contribution with increasing α . Upon further increasing the damping, however, the temporal weight decreases significantly and reaches a substantially smaller value at $\alpha = 0.2$. This decrease reflects the increasingly strong dissipation of the coherent exchange oscillation, which limits its temporal persistence and suppresses the nonlinear response. The non-monotonic dependence of the temporal weight therefore provides a quantitative indication of the competition between the damping-enhanced FDT contribution and the damping-induced suppression of coherent exchange dynamics. The maximum around $\alpha \approx 0.05$ should be regarded as specific to the model parameters and excitation conditions considered here rather than as a universal optimum damping value.

The damping value $\alpha \approx 0.01$ identified in these simulations should not be interpreted as a universal threshold for the observability of FDT. Rather, it represents an approximate crossover for the specific two-sublattice model, exchange-mode frequency, sublattice pa-

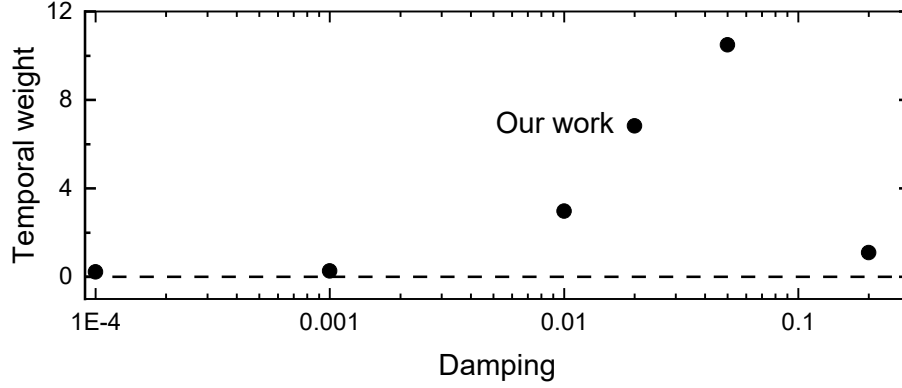


Figure 5.6: Temporal weight of the exchange oscillation as a function of the Gilbert damping parameter.

rameters, and THz excitation conditions considered in this study. The relative importance of FDT depends not only on the Gilbert damping parameter but also on the excitation frequency, sublattice volume per spin, gyromagnetic ratios, exchange interactions, and the ability of the system to sustain coherent exchange dynamics. Thus, the value $\alpha \approx 0.01$ should be regarded as a model- and parameter-dependent criterion for the onset of appreciable FDT-induced nonlinear features under the present conditions, rather than as a universal material threshold.

5.3 Implications of FDT for THz magnetization reversal

The understanding gained from the material-dependent behavior of the field-derivative torque also provides a promising route towards ultrafast magnetization reversal in ferromagnetic systems. Building upon the conditions identified in this chapter, recent numerical investigations on GdYb-BIG have shown that the inclusion of FDT substantially lowers the THz magnetic field required for deterministic magnetization switching under circularly polarized THz excitation [106]. The additional relativistic torque enhances the effective driving field experienced by the magnetic sublattices, enabling coherent reversal at significantly lower field amplitudes compared to the conventional Zeeman torque alone. Fur-

thermore, the switching phase diagram reveals a considerable expansion of the switching regime in the presence of FDT, demonstrating that the material parameters governing the strength of FDT not only influence nonlinear magnetization dynamics but also play a crucial role in reducing the energy cost of ultrafast spin reversal [107]. These findings further establish high-damping rare-earth ferrimagnets as promising candidates for energy-efficient THz spintronic and magnetic memory applications.

5.4 Summary

In this chapter, we have presented a comprehensive theoretical investigation of the material-dependent aspects of field-derivative-torque-driven nonlinear magnetization dynamics in ferrimagnetic systems, complementing the experimental demonstration of Chapter 4. Using numerically solved coupled FDT-modified LLG equations, we first examined the linear response of the ferrimagnetic system under single-pulse THz excitation and demonstrated that the inclusion of FDT leads to a substantial enhancement of the Kaplan-Kittel exchange mode amplitude alongside a frequency-dependent phase shift in the effective driving field – effects that are absent in the conventional Zeeman-torque-only description. Extending the analysis to dual-pulse nonlinear excitation, we showed that the sublattice spin-volume ratio $a_{\text{Fe}}^3/a_{\text{RE}}^3$ plays a decisive role in governing both the amplitude and phase of the FDT-induced Néel-vector oscillations, with a characteristic transition in the phase shift occurring near the antiferromagnetic limit where the two sublattice volumes are equal. Most significantly, these observations highlight the dual role of Gilbert damping in nonlinear magnetization dynamics. Increasing α enhances the relative strength of the FDT contribution; however, sufficiently large damping also increases dissipation and suppresses the coherent exchange dynamics required to resolve the nonlinear response. Consequently, the emergence of appreciable FDT-induced nonlinear features and their subsequent suppres-

sion at high damping reflect a competition between damping-enhanced relativistic driving and damping-induced loss of coherent spin dynamics. The resulting crossover and upper damping limit are specific to the material parameters and excitation conditions considered in the present model and should not be interpreted as universal damping thresholds. An upper damping limit was also identified, above which the exchange magnon mode bifurcates and the nonlinear spectrum becomes incoherent. These results provide a quantitative explanation for why FDT was not observed in previously studied low-damping systems such as YFeO_3 and $\text{Tm}_2\text{BiFe}_{4.2}\text{Ga}_{0.8}\text{O}_{12}$, and establish concrete material design criteria, high Gilbert damping, a THz-frequency exchange resonance, equivalent sublattice gyromagnetic ratios, and a sublattice spin-volume ratio away from unity, for identifying future ferrimagnetic systems in which relativistic spin-torque effects can be accessed and exploited at THz frequencies. Note that the material-dependent trends discussed above should be regarded as general physical guidelines rather than universal quantitative criteria. The present analysis is based on a two-sublattice model consisting of rare-earth and Fe sublattices with different magnetic moments and antiferromagnetic inter-sublattice coupling, as appropriate for the ferrimagnetic system considered in this thesis. The same two-sublattice framework can also be applied to a compensated antiferromagnet, although the vanishing or strongly suppressed net magnetization makes dynamics of the individual sublattices less directly accessible to probes that primarily couple to the net magnetization. Within the present framework, a conventional ferromagnet, in which the magnetic dynamics can be represented by a single effective magnetic sublattice, does not exhibit the same two-sublattice FDT dynamics considered here. The two-sublattice description can also be extended to systems in which additional magnetic sublattices contribute significantly to the dynamics. This is particularly relevant to rare-earth iron garnets containing more than one magnetically active rare-earth species. In such cases, a three-sublattice or more general multi-sublattice description may be required, with the FDT contribution evaluated for each relevant sublattice using its cor-

responding magnetic moment, gyromagnetic ratio, damping parameter, and volume per spin. The additional coupled degrees of freedom can modify the quantitative conditions for the observability of FDT-induced nonlinear dynamics. The model can likewise be extended to include exchange interactions beyond the nearest-neighbor approximation, such as next-nearest-neighbor interactions, when these interactions are relevant to the magnetic dynamics [44].

Chapter 6

The interplay of crystal-field transitions and exchange spin dynamics in a ferrimagnet

The preceding two chapters established that the nonlinear THz magnetization dynamics of GdYb-BIG cannot be described by conventional Zeeman torque alone, and that the relativistic field-derivative torque is essential for reproducing the experimentally observed $\chi^{(3)}$ nonlinear signals. These investigations were carried out at room temperature, where the ferrimagnetic order is well established, the Kaplan-Kittel exchange mode is sharp and well-defined, and the rare-earth sublattice is dominated by the Gd^{3+} contribution. However, as the temperature is lowered toward and below the magnetization compensation temperature $T_{\text{MC}} = 96 \text{ K}$, an entirely new aspect of the spin dynamics of GdYb-BIG comes into play, the Yb^{3+} sublattice magnetization, which is negligibly small at room temperature, grows rapidly and begins to compete with and ultimately dominate the rare-earth sublattice contribution. Simultaneously, the strong spin-orbit coupling of the Yb^{3+} ion gives rise to crystal-electric-field (CEF) excitations within the THz frequency range that are directly accessible to THz spectroscopy and, as demonstrated in this chapter, hybridize strongly with the collective Yb-Fe exchange resonance. This hybridization, and its unexpected spectroscopic consequence – an anomalous redshift of the exchange mode upon cooling that contradicts the conventional expectation of a blue-shift driven by strengthening exchange, constitutes the primary result of the present chapter.

6.1 Crystal electric field

In a free ion, the electronic energy levels are determined by the intra-atomic Coulomb interaction and spin-orbit coupling, and the $2J + 1$ states of a given J multiplet are degenerate in the absence of any external perturbation. In a crystalline solid, however, this degeneracy is partially or fully lifted by the electrostatic potential generated by the surrounding lattice of neighbouring ions – the crystal electric field (CEF) [52]. The CEF arises because the electrons of the magnetic ion are no longer in a spherically symmetric environment, instead, they experience the non-uniform electrostatic potential produced by the charged ligand ions that surround the magnetic site. This potential, sometimes referred to as the crystalline electric field or crystal field, acts on the orbital degree of freedom of the unpaired electrons and redistributes the energy of the otherwise degenerate m_J states according to the local point-group symmetry of the magnetic site. The resulting splitting of the J multiplet into a set of lower-symmetry energy levels is known as crystal-field splitting. For transition-metal ions, where the relevant $3d$ orbitals extend spatially into the ligand environment, the crystal-field splitting is typically large, on the order of 10^3 - 10^4 cm^{-1} and constitutes the dominant perturbation after the intra-atomic Coulomb interaction. For rare-earth ions, by contrast, the magnetic $4f$ electrons are spatially localized well within the outer closed shells and are therefore comparatively well shielded from the crystalline environment. The CEF splitting of rare-earth J multiplets is correspondingly smaller, of order 10^2 cm^{-1} , placing the lowest CEF transitions in the meV energy range. An important consequence of the crystal-field interaction for rare-earth ions is Kramer's theorem: for ions with an odd number of electrons, time-reversal symmetry guarantees that every energy level remains at least twofold degenerate regardless of the crystal symmetry, in the absence of a magnetic field [52]. In GdYb-BIG, Yb³⁺ CEF transition at approximately 0.7 THz makes it directly relevant to the temperature-dependent THz dynamics reported in this chapter.

6.2 Electronic structure of Yb^{3+} in GdYb-BIG

A quantitative understanding of the CEF excitations in GdYb-BIG begins with the electronic configuration of the Yb^{3+} ion. The $4f^{13}$ ground state of Yb^{3+} has a single unpaired electron in an otherwise filled $4f$ shell. Strong spin-orbit coupling, which is the dominant electronic interaction for $4f$ electrons, couples the orbital ($L = 3$) and spin ($S = 1/2$) angular momenta into total angular momentum multiplets $J = L + S = 7/2$ and $J = L - S = 5/2$, separated by a spin-orbit splitting of approximately 1.3 eV, see Figure 6.1a. At the low energies relevant for the magnetization dynamics studied in this thesis, only the $J = 7/2$ ground state multiplet is thermally populated.

Within the $J = 7/2$ multiplet, the surrounding crystalline electrostatic potential from the O^{2-} ligand ions at the dodecahedral site of the garnet structure further lifts the eight-fold degeneracy (corresponding to $m_J = -7/2, -5/2, \dots, +7/2$) into a set of Kramers doublets, since Yb^{3+} is a half-integer spin system and time-reversal symmetry guarantees at least twofold degeneracy in the absence of a magnetic field. This CEF splitting results in doublets with energy separations that fall in the meV range, resonant with THz frequencies. An additional splitting of these doublets is induced by the internal exchange field of the Fe^{3+} sublattice, which acts on the Yb^{3+} moment and attempts to align it antiparallel to the net Fe magnetization [50, 51]. This exchange-field-induced final splitting of the lowest doublet produces energy levels with a spacing of approximately 0.7 THz in GdYb-BIG, as shown schematically in Figure 6.1a, placing the lowest CEF transition directly within the spectral window of our THz time-domain spectroscopy measurements.

The key distinction between Gd^{3+} and Yb^{3+} in this context is important. The Gd^{3+} ion has a half-filled $4f^7$ shell with $L = 0$ and $S = 7/2$, giving it zero orbital angular momentum. As a result, the spin-orbit coupling of Gd^{3+} is negligibly small, and it does not support CEF transitions in the THz range. The Yb^{3+} ion, by contrast, carries a large orbital moment

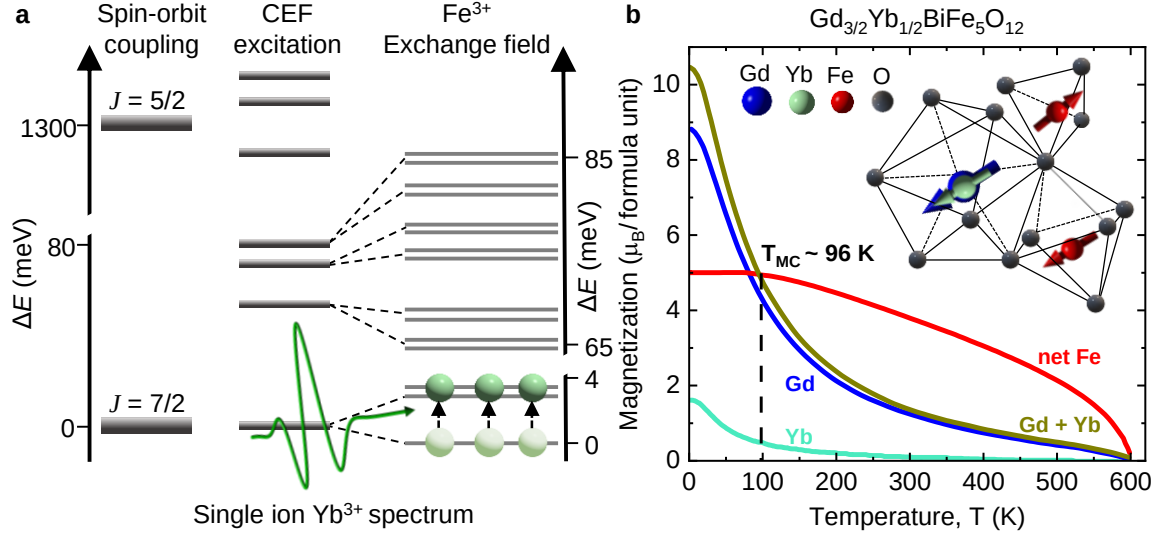


Figure 6.1: **(a)** Electronic energy-level scheme of the Yb^{3+} ($4f^{13}$) ion. The ground-state multiplet is split by the combined effects of spin-orbit coupling, the crystal electric field (CEF), and the exchange field arising from the Fe^{3+} sublattice. The lowest-lying CEF transitions fall within the experimentally accessible THz frequency range. **(b)** Temperature dependence of the rare-earth (RE) and Fe sublattice magnetizations in $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ (adapted from Figure 3.15 of Ref. [11]). At the magnetic compensation temperature (T_{MC}), the net magnetization of the RE sublattice exactly compensates that of the Fe sublattice, resulting in a vanishing total magnetization.

($L = 3$), making it strongly susceptible to the crystalline electric field and giving rise to the THz-frequency CEF excitations that are central to the results of this chapter. Furthermore, the non-zero orbital angular momentum of Yb^{3+} modifies its effective gyromagnetic ratio and makes the Yb-Fe exchange interaction intrinsically anisotropic at low temperatures. As discussed in Section 6.6, this modification in the Yb-Fe exchange energy is responsible for the anomalous redshift of the Yb-Fe exchange resonance.

6.3 Temperature-dependent sublattice magnetization

Having established the electronic structure of the Yb^{3+} ion and the origin of its THz-frequency CEF excitations, we now examine how the sublattice magnetizations of $\text{GdYb-BiFe}_5\text{O}_{12}$ evolve with temperature, since it is this thermal evolution that drives the progres-

sive emergence of the CEF-mediated dynamics observed in the THz measurements. Figure 6.1b shows the temperature-dependent sublattice magnetizations of GdYb-BIG, calculated within molecular field theory [1]. At room temperature, the net rare-earth moment is dominated by the Gd^{3+} contribution, since the Gd sublattice magnetization is already well developed at this temperature while the Yb^{3+} moment remains negligibly small. As the temperature is reduced below approximately 150 K, the Yb^{3+} sublattice magnetization begins to develop appreciably, growing steadily as thermal fluctuations are progressively suppressed and the population of the lowest CEF doublet increases. This onset of the Yb moment is entirely consistent with the electronic picture of the preceding section: only as the system is cooled sufficiently below the CEF splitting energy does the Yb^{3+} ground-state magnetization become thermally resolvable. The Yb contribution increases sharply at cryogenic temperatures and plays an increasingly important role in the total rare-earth sublattice moment at low temperatures. At the magnetization compensation temperature $T_{\text{MC}} = 96$ K, the net rare-earth moment, now with a significant Yb^{3+} contribution, becomes equal in magnitude and opposite in direction to the Fe sublattice moment, causing the net macroscopic magnetization to vanish. Below T_{MC} , the rare-earth contribution dominates and the sign of the net magnetization reverses. This temperature window, in which the Yb^{3+} moment is active and the system passes through its compensation point, is precisely the regime in which the CEF-exchange hybridization reported in the following section is most pronounced.

6.4 Temperature-dependent THz-TDS

To investigate the temperature-dependent spin dynamics of GdYb-BIG, we perform THz time-domain spectroscopy (THz-TDS) in the transmission geometry across a temperature range from 10 K to 170 K, spanning the magnetization compensation temperature

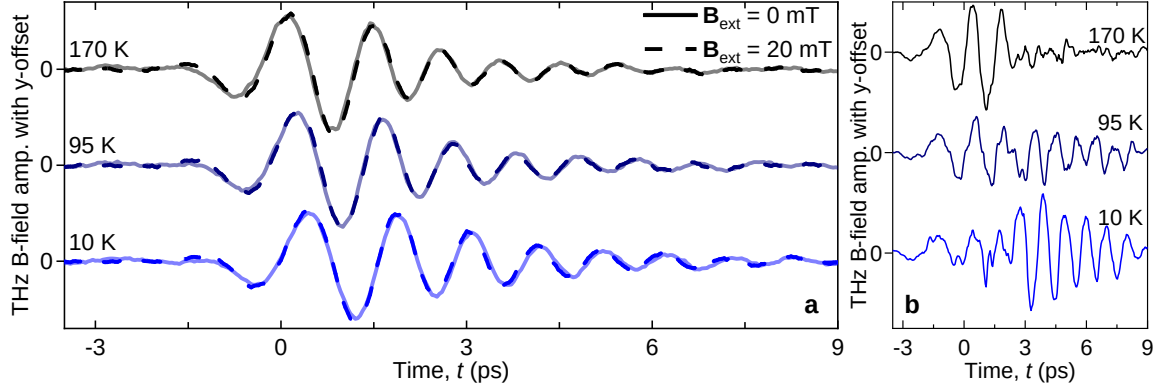


Figure 6.2: **(a)** Temperature-dependent THz time-domain transients of $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ measured in the absence (dashed lines) and presence (solid lines) of an in-plane external magnetic field of 20 mT. The oscillatory component observed under the applied magnetic field primarily arises from the intrinsic THz absorption of the material within the measured frequency range. **(b)** Difference signal obtained by subtracting the zero-field response from the finite-field response at each temperature, revealing low- and high-frequency oscillatory components with distinctly different temperature evolutions.

$T_{\text{MC}} = 96 \text{ K}$. All measurements are performed in the presence of a small in-plane external magnetic field of $\mathbf{B}_{\text{ext}} = 20 \text{ mT}$. To isolate the field-induced magnetic response from the dominant background absorption of the material which noted in Chapter 4, the time transients recorded at $\mathbf{B}_{\text{ext}} = 20 \text{ mT}$ are subtracted from the corresponding zero-field measurements, see Figure 6.2. The differential signals therefore reflect exclusively the field-induced changes in the magnetic response of the system.

6.4.1 Temperature evolution of the time transients

The temperature-dependent differential THz transients are shown in Figure 6.3a. At high temperatures (e.g., 170 K), the response is dominated by a single low-frequency (LF) oscillatory component. As the temperature is lowered toward T_{MC} , this LF component progressively attenuates, while a new high-frequency (HF) oscillatory component emerges and strengthens. Strikingly, near $T_{\text{MC}} \approx 96 \text{ K}$, the LF and HF components attain comparable amplitudes, and a pronounced redistribution of spectral weight between them is observed.

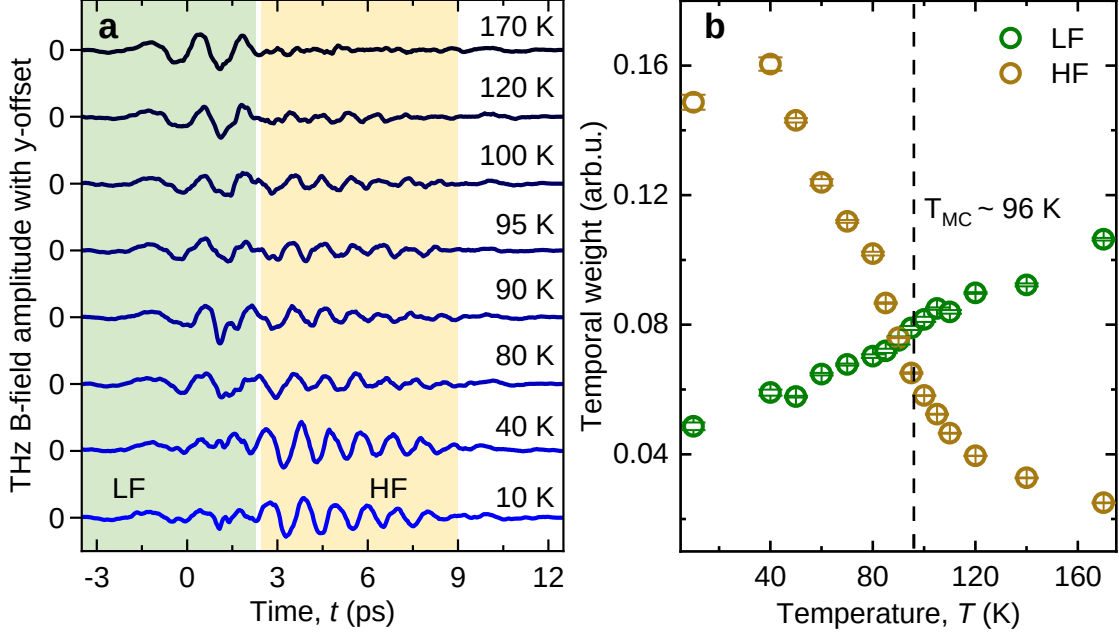


Figure 6.3: **(a)** Temperature-dependent time transients of GdYb-BIG, recorded in the presence of an external in-plane magnetic field of 20 mT. The time transients reveal low-frequency (LF) and high-frequency (HF) oscillatory components, denoted by the green- and yellow-shaded regions, respectively. As the LF component decreases, the HF component begins to develop below a certain temperature and strengthens thereafter. **(b)** The temperature-dependent spectral weights associated with the LF and HF components cross near the magnetization compensation temperature (T_{MC}), featuring a cross-talk characteristic of the coupled dynamics.

Below T_{MC} , the HF component continues to grow while the LF component further diminishes. This temperature evolution of the two oscillatory components provides the first clear experimental evidence of a coupled, hybridized dynamics in the system.

To quantify this redistribution, we compute the temporal weight associated with each component by integrating the squared amplitude of each oscillation over the time window marked by the green and yellow shaded region in Figure 6.3a. As shown in Figure 6.3b, the temporal weights of the LF and HF components are found to cross near T_{MC} , exhibiting a characteristic anti-crossing behaviour that is a hallmark of strongly coupled modes exchanging spectral weight.

6.4.2 Frequency-domain analysis and mode assignment

To identify the frequency components of the LF and HF oscillations, we apply a fast Fourier transform to the temperature-dependent time transients. The resulting temperature-dependent spectra, shown in Figure 6.4a, reveal three distinct resonance peaks across the measurement temperature range. The first, appearing near 0.3 THz and remaining relatively temperature-independent in frequency, is tentatively attributed to the Gd-Fe exchange dynamics, since the Gd^{3+} sublattice magnetization evolves gradually with temperature and its exchange coupling to Fe sets a mode at a frequency governed by $\lambda_{\text{Gd-Fe}}$. The second resonance, centered near 0.7 THz and developing prominently below approximately 150 K, is assigned to the CEF excitation of the Yb^{3+} single ion: its onset temperature is consistent with the progressive occupation of the lower CEF doublet as thermal fluctuations are suppressed upon cooling, and its frequency matches the 0.7 THz CEF splitting identified from the electronic structure. The third resonance, appearing near 1 THz at low temperatures, tracks the Yb sublattice magnetization in its temperature dependence, growing as the Yb moment increases upon cooling, and is assigned to the Yb-Fe exchange dynamics.

To extract quantitative mode parameters, each spectrum is fitted with a sum of Lorentzian profiles. The temperature dependencies of the spectral weight, linewidth, and resonance frequency of the CEF and Yb-Fe exchange modes, extracted from these fits, are shown in Figures 6.4b, c, and d respectively. Three observations are noteworthy. First, the spectral weights of the 0.7 THz CEF mode and the 1 THz Yb-Fe exchange mode exhibit a pronounced crossing near T_{MC} , consistent with the temporal weight analysis in the time domain. Second, while the linewidth of the CEF excitation narrows upon cooling – as expected from the suppression of thermal broadening at low temperatures, the linewidth of the Yb-Fe exchange mode remains remarkably constant across the entire temperature

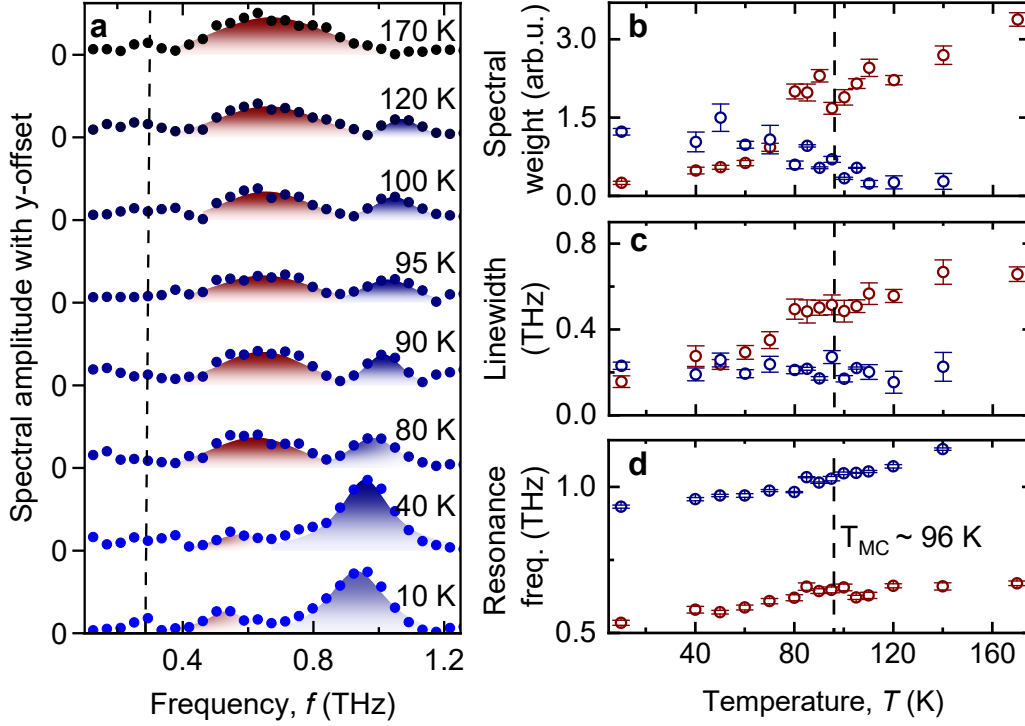


Figure 6.4: **(a)** Temperature-dependent spectra of the time transients in Figure 6.3a. The spectra reveal the temperature evolution of the individual modes, namely, the Gd-Fe exchange dynamics (at 0.3 THz), the Yb-ion CEF excitation (at 0.7 THz), and the Yb-Fe exchange dynamics (at 1 THz). The red- and blue-shaded regions represent the corresponding Lorentz fit of the CEF excitation and Yb-Fe exchange frequency components. **(b)** The spectral weight of the CEF excitation and Yb-Fe exchange mode shows a redistribution near the magnetization compensation point. The temperature-dependent **(c)** linewidth and **(d)** the resonance frequency of these two modes, respectively. While the CEF linewidth narrows with decreasing temperature, the Yb-Fe exchange mode linewidth remains nearly unchanged. Moreover, both resonance modes exhibit a redshift in their frequencies as the temperature decreases.

range. This temperature-independent linewidth indicates that the Yb-Fe exchange precession is a predominantly coherent, homogeneously broadened mode throughout. Third, and most strikingly, both the 0.7 THz CEF resonance and the 1 THz Yb-Fe exchange resonance exhibit a pronounced redshift upon cooling, as clearly visible in Figure 6.4d.

6.5 An estimate of the CEF-exchange coupling

The temperature-dependent resonance frequencies in Figure 6.4d provide a quantitative estimate of the coupling between the localized Yb-CEF excitation and the collective Yb-Fe exchange mode. The two modes form coupled branches as their frequencies approach each other near the magnetization compensation regime. Within a simple coupled-oscillator description, the eigen-frequencies of two modes coupled with strength g exhibit an avoided crossing, with the minimum frequency splitting given by

$$\Delta\omega_{\min} = 2g.$$

Expressed in terms of ordinary frequency $f = \omega/2\pi$, this relation becomes

$$\Delta f_{\min} = 2 \frac{g}{2\pi}.$$

From the resonance frequencies extracted from Figure 6.4d, the separation between the two hybridized branches near $T \approx 95$ K is approximately 0.38 THz. This gives an effective coupling strength of

$$\frac{g}{2\pi} \approx \frac{\Delta f_{\min}}{2} \approx 0.19 \text{ THz}.$$

This coupling strength is comparable to the linewidths of the two modes in the vicinity of the hybridization regime and is therefore sufficiently large to produce appreciable mode mixing. The estimated coupling provides a quantitative characterization of the CEF–exchange hybridization and is consistent with the pronounced redistribution of spectral weight between the Yb-CEF and Yb-Fe exchange modes observed near T_{MC} .

6.5.1 The anomalous redshift

The temperature dependence of the resonance frequency shown in Figure 6.4d constitutes the most unexpected finding of this chapter. In a conventional ferrimagnet, the exchange

resonance frequency is expected to increase upon cooling as the exchange constants strengthen with decreasing temperature and the sublattice magnetizations grow toward their saturation values. This is the standard blue-shift of exchange-driven modes, observed in the vast majority of magnetic systems. In GdYb-BIG, the Yb-Fe exchange mode behaves in precisely the opposite way: its resonance frequency decreases systematically as the temperature is lowered from 170 K to 10 K. The CEF excitation at 0.7 THz also softens upon cooling, tracking the redshift of the Yb-Fe mode. This co-redshift of both the CEF and exchange resonances is a direct signature of their mutual coupling: the two modes are not independent resonances that happen to coexist in the same spectral window, but are hybridized excitations whose frequencies are jointly renormalized by the same microscopic interaction.

6.6 Theoretical modeling: Three-sublattice LLG

To elucidate the microscopic origin of the anomalous redshift and the CEF-exchange hybridization, we extend the two-sublattice LLG framework of Chapters 2 and 4 to a three-sublattice model that explicitly accounts for the distinct thermal and dynamical behaviors of the Gd and Yb rare-earth sublattices. The equation of motion for each sublattice i ($=$ Fe, Gd, Yb) remains of the standard LLG form, Eq. (4.7), with sublattice-dependent gyromagnetic ratios γ_i , damping parameters α_i , and effective fields $\mathbf{B}_i^{\text{eff}} = -\delta\mathcal{F}/\delta\mathbf{m}_i$.

6.6.1 Three-sublattice free energy

The free energy of the three-sublattice system is [108–110]

$$\begin{aligned}
 \mathcal{F}(\mathbf{m}_{\text{Fe}}, \mathbf{m}_{\text{Gd}}, \mathbf{m}_{\text{Yb}}) = & -\lambda_{\text{Gd-Fe}} \mathbf{m}_{\text{Gd}} \cdot \mathbf{m}_{\text{Fe}} - \lambda_{\text{Yb-Fe}} \mathbf{m}_{\text{Yb}} \cdot \mathbf{m}_{\text{Fe}} - \lambda_{\text{Gd-Yb}} \mathbf{m}_{\text{Gd}} \cdot \mathbf{m}_{\text{Yb}} \\
 & - K_{\text{Fe}} \frac{(\mathbf{m}_{\text{Fe}} \cdot \mathbf{n})^2}{|\mathbf{m}_{\text{Fe}}|^2} - K_{\text{Gd}} \frac{(\mathbf{m}_{\text{Gd}} \cdot \mathbf{n})^2}{|\mathbf{m}_{\text{Gd}}|^2} - K_{\text{Yb}} \frac{(\mathbf{m}_{\text{Yb}} \cdot \mathbf{n})^2}{|\mathbf{m}_{\text{Yb}}|^2} \\
 & - [\mathbf{B}_{\text{THz}}(t) + \mathbf{B}_{\text{ext}}] \cdot (\mathbf{m}_{\text{Fe}} + \mathbf{m}_{\text{Gd}} + \mathbf{m}_{\text{Yb}}) \\
 & + \frac{\mu_0}{2} (\mathbf{m}_{\text{Fe}} \cdot \mathbf{n} + \mathbf{m}_{\text{Gd}} \cdot \mathbf{n} + \mathbf{m}_{\text{Yb}} \cdot \mathbf{n})^2.
 \end{aligned} \tag{6.1}$$

where $\lambda_{\text{Gd-Fe}}$, $\lambda_{\text{Yb-Fe}}$, and $\lambda_{\text{Gd-Yb}}$ are the inter-sublattice exchange constants, and K_{Fe} , K_{Gd} , K_{Yb} are the uniaxial anisotropy constants for each sublattice. The coupling between the Gd and Yb sublattices ($\lambda_{\text{Gd-Yb}}$) is ferromagnetic in character, while significantly smaller than the antiferromagnetic Fe-RE couplings, exerts a discernible influence on the global frequency response and must be included for quantitative agreement. The three-sublattice model reproduces the three spectral resonances: the 0.3 THz Gd-Fe exchange mode, dominated by the increasing Gd-Fe exchange energy upon cooling; the 0.7 THz Yb-CEF mode; and the 1 THz Yb-Fe exchange mode.

6.6.2 Anisotropic Yb-Fe exchange and the origin of the redshift

The key physical picture responsible for the anomalous redshift is the temperature dependence of the Yb-Fe exchange interaction. Unlike the Gd-Fe coupling, which is well approximated as isotropic and strengthens monotonically upon cooling, the Yb-Fe exchange interaction becomes strongly anisotropic at low temperatures as a consequence of the non-zero orbital angular momentum of Yb^{3+} , its strong spin-orbit coupling, and the progressive occupation of the lower CEF levels [111]. The CEF modifies the direction-dependent response of the Yb^{3+} moment and consequently the exchange interaction between the Yb and Fe sublattices. This Yb-Fe exchange is mediated through the oxygen-containing superexchange pathways in the garnet structure. The resulting effective anisotropic exchange

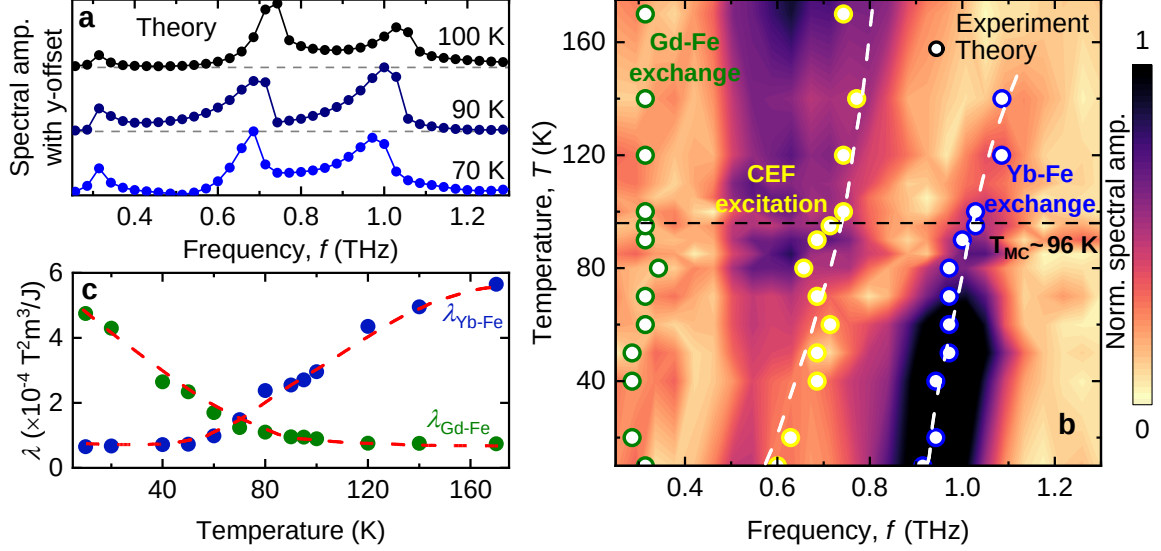


Figure 6.5: **(a)** Theoretical resonance spectra at three representative temperatures, identifying the three primary modes: Gd-Fe exchange, Yb-ion crystal electric field (CEF) excitation, and Yb-Fe exchange. **(b)** Contour plot of the temperature-dependent experimental spectra. The resonance frequencies derived from our theoretical model (open circles) exhibit excellent agreement with the experimental data, highlighting the hybridization between the Yb-ion CEF excitation and Yb-Fe exchange dynamics. White dashed lines are provided as a guide to the eye. **(c)** Temperature dependence of the Yb-Fe and Gd-Fe exchange energy. The reduction in Yb-Fe exchange energy at lower temperatures drives a pronounced red-shift in both the Yb-ion CEF and Yb-Fe exchange resonance modes. The red-dashed line is a guide to the eye.

interaction can be represented by the tensor

$$\lambda_{\text{Yb-Fe}} = \begin{pmatrix} 2.26\lambda & 3.30\lambda & -0.47\lambda \\ -1.91\lambda & -2.87\lambda & 3.22\lambda \\ 4.04\lambda & 4.68\lambda & -2.24\lambda \end{pmatrix}, \quad (6.2)$$

where λ is a scalar, temperature-dependent variable. The numerical coefficients in Eq. (6.2) are not introduced as phenomenological fitting parameters to reproduce the temperature-dependent THz spectra. Instead, they arise from the transformation of the anisotropic Yb-Fe exchange interaction from the crystallographic coordinate system to the local coordinate system of the Yb ion. To describe the local crystal-field environment, the exchange interaction is formulated using Wybourne operators, whose principal axes are defined with respect to the local symmetry of the Yb site. Following the local-coordinate transformation

procedure reported in, the crystallographic basis of the Yb sites is transformed into the corresponding local basis. Applying this transformation to the present garnet structure gives the numerical tensor coefficients appearing in Eq. (6.2). Thus, the tensor provides an effective representation of the direction-dependent $4f$ - $3d$ exchange interaction in the three-sublattice LLG model rather than an independently fitted tensor constrained by the present THz spectra. The off-diagonal elements of the tensor are particularly important because they couple different magnetization components and therefore retain the CEF-induced anisotropy of the Yb-Fe exchange interaction. As a numerical check, suppressing these off-diagonal elements substantially modifies the hybridized resonance structure, demonstrating that they are essential for describing the coupled Yb-CEF and Yb-Fe exchange dynamics. As the temperature decreases, the preferential occupation of the lowest CEF doublet enhances the net Yb sublattice magnetization and simultaneously reduces the effective isotropic component of the Yb-Fe exchange coupling - since the CEF-driven anisotropy partially counteracts the exchange-field-induced alignment. The net consequence is that the effective Yb-Fe exchange energy $\lambda_{\text{Yb-Fe}}$ decreases with decreasing temperature, as shown in Figure 6.5c, in contrast to the Gd-Fe exchange energy which increases. This temperature-dependent reduction of the effective Yb-Fe coupling is directly responsible for the observed redshift of both the Yb-Fe exchange mode and the Yb-CEF excitation, since both modes are governed primarily by the Yb-Fe interaction strength.

Physically, this mechanism can be understood as follows. Near room temperature, thermal agitation distributes the Yb^{3+} population nearly uniformly across the CEF-split energy levels, rendering the Yb^{3+} ions effectively isotropic and magnetically inactive. As the temperature decreases below approximately 150 K, thermal fluctuations are progressively suppressed and the Yb^{3+} population concentrates in the lowest CEF doublet. The non-zero orbital angular momentum of Yb^{3+} in this lowest state makes it energetically non-equivalent to the Fe^{3+} ions, introducing a highly anisotropic exchange interaction $\lambda_{\text{Yb-Fe}}$ between the

Yb and Fe sublattices [112]. The net effect of this anisotropic exchange, mediated by the competing influence of the Fe exchange field and the CEF manifold through the spin-orbit coupling – is a reduction of the effective isotropic Yb-Fe exchange energy. This drives the observed redshift of the exchange resonance upon cooling. This constitutes a CEF-mediated renormalization of the Yb single-ion anisotropy and the effective exchange coupling – an effect entirely absent for the orbital-moment-free Gd^{3+} sublattice.

6.6.3 Agreement between theory and experiment

The three-sublattice LLG model with the anisotropic, temperature-dependent Yb-Fe exchange tensor is solved numerically for the temperature-dependent THz response. The theoretical resonance spectra at three representative temperatures (70 K, 90 K, and 100 K) are shown in Figure 6.5a, reproducing the three experimentally observed modes at approximately 0.3 THz, 0.7 THz, and 1 THz with good qualitative and quantitative agreement. The temperature dependence of the resonance frequencies extracted from the model is overlaid on the experimental data in Figure 6.5b as open circles, showing excellent agreement across the full temperature range from 10 K to 170 K. In particular, the model correctly reproduces the anomalous redshift of both the Yb-CEF and Yb-Fe exchange modes upon cooling, as well as the relative stability of the Gd-Fe mode frequency. The temperature dependence of $\lambda_{\text{Yb-Fe}}$ and $\lambda_{\text{Gd-Fe}}$ is shown in Figure 6.5c, confirming the physical picture: the Gd-Fe exchange strengthens monotonically upon cooling, while the effective Yb-Fe exchange weakens, driving the redshift. The full temperature dependent spectra, obtained from the simulation is shown in Figure 6.6.

6.7 Summary

In this chapter, we have investigated the temperature-dependent THz magnetization dynamics of GdYb-BIG across its magnetic compensation temperature $T_{\text{MC}} = 96$ K, revealing a

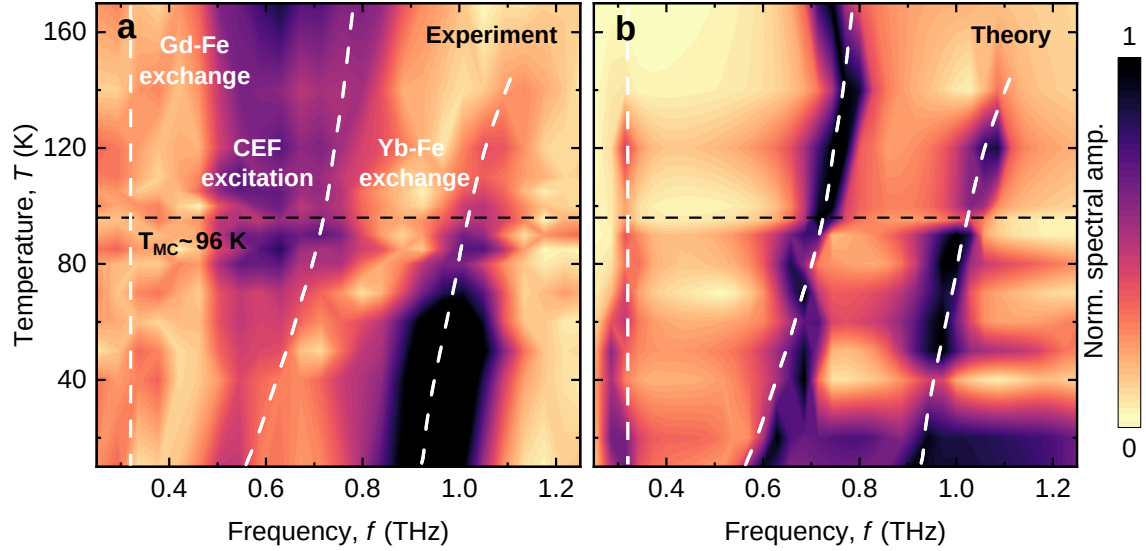


Figure 6.6: **(a)**, **(b)** Temperature-dependent 2D contour plot of the experimental and theoretically obtained spectra of GdYb-BIG, respectively. The theoretical spectra, obtained from the three-sublattice model, corroborate the experimental results and reveal a strong hybridization between the localized Yb-ion crystal-electric-field (CEF) excitation and the Yb-Fe exchange interaction. The white dashed lines are the guide-to-the-eye, highlighting the temperature evolution of the coupled resonances.

rich interplay between localized Yb^{3+} crystal-electric-field excitations and collective exchange spin dynamics. Temperature-dependent THz time-domain spectroscopy identified three spectral modes: a Gd-Fe exchange mode near 0.3 THz, a Yb^{3+} CEF excitation near 0.7 THz, and a Yb-Fe exchange mode near 1 THz. A pronounced redistribution of spectral and temporal weight between the CEF and Yb-Fe exchange modes near T_{MC} , exhibiting a characteristic anti-crossing behavior, provided direct evidence of strong hybridization between these two excitations. Most strikingly, both the CEF and Yb-Fe exchange resonances were found to redshift upon cooling – the opposite of the blue-shift conventionally expected from strengthening exchange interactions, revealing an anomalous softening of the exchange dynamics at low temperatures.

A three-sublattice LLG model incorporating an anisotropic, temperature-dependent Yb-Fe exchange tensor, whose physical origin lies in the CEF-driven renormalization of the Yb

single-ion anisotropy mediated by spin-orbit coupling, was shown to quantitatively reproduce both the three-mode structure and the anomalous redshift across the full temperature range. The physical picture that emerges is one in which the progressive population of low-energy CEF doublets upon cooling increases the anisotropy of the Yb-Fe exchange interaction while simultaneously reducing its effective isotropic component, thereby softening rather than hardening the Yb-related exchange dynamics. These findings establish that the magnetic anisotropy of a rare-earth ferrimagnetic garnet is a dynamically renormalized, temperature-dependent quantity governed by the CEF level structure, and they demonstrate that rare-earth iron garnets constitute a versatile platform for investigating hybridized orbital-spin dynamics in the THz frequency range. The broader implications of these results for the design and control of THz-frequency spintronic and magnonic systems are discussed in the concluding chapter.

Chapter 7

Summary and Outlook

7.1 Summary

The primary objective of this thesis was to address a fundamental question on relativistic effects in ultrafast magnetism: Is the conventional LLG equation sufficient to describe magnetization dynamics on ultrafast timescales, or do additional relativistic corrections become important? Although the LLG framework based on Zeeman torque has been remarkably successful in explaining a wide range of magnetic phenomena, theoretical studies have suggested that spin-orbit coupling, the relativistic interaction responsible for Gilbert damping, can generate an additional torque on the magnetization when the driving magnetic field changes rapidly in time. This so-called field-derivative torque had remained a purely theoretical prediction, with no direct experimental evidence of its existence. Determining whether this relativistic correction leaves a measurable signature in the spin dynamics of a material driven by intense THz pulses, and identifying the material conditions under which such signatures become experimentally observable, formed the primary motivation of this thesis.

In this regard, the rare-earth iron garnet, $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$ proved to be an ideal platform. Chapter 4 presented the experimental evidence of the FDT in the nonlinear spin dynamics of the garnet. By combining THz time-domain spectroscopy, THz-pump optical-probe Faraday studies, and nonlinear two-dimensional THz spectroscopy, the Kaplan-Kittel exchange mode of GdYb-BIG was identified at 0.48 THz and then resonantly driven into a nonlinear regime. The observed nonlinear spectrum of the exchange dynamics consisted of four distinct $\chi^{(3)}$ signals, which could not be reproduced within the conventional LLG

framework. Remarkably, incorporating the FDT into the theoretical model successfully reproduced all experimentally observed nonlinear features, providing direct evidence of the importance of relativistic effects in ultrafast magnetization dynamics.

Chapter 5 addressed a natural question arising from these observations: Why is the effect of FDT visible in GdYb-BIG but absent in many other magnetic systems? Through systematic numerical simulations, it was shown that the strength and phase of the FDT contribution are governed primarily by the Gilbert damping and the ratio of the unit-cell volumes per spin of the individual magnetic sublattices. A threshold damping value was identified, below which the FDT remains too weak to generate observable signatures. In contrast, the relatively large damping of GdYb-BIG makes the relativistic contribution sufficiently strong to produce distinct nonlinear responses. The study also revealed an upper damping limit beyond which the exchange mode loses coherence. These results provide useful guidelines for identifying other materials in which relativistic spin torques may become experimentally accessible. Furthermore, it was found that the presence of the FDT lowers the THz peak field required to induce magnetization precessional switching in the same garnet system, highlighting its potential for energy-efficient ultrafast spin manipulation.

Finally, Chapter 6 explored the temperature evolution of the exchange dynamics in GdYb-BIG across its magnetic compensation point. Temperature-dependent THz spectroscopy revealed a hybridization between the Yb single-ion CEF excitation and Yb-Fe exchange mode. Unexpectedly, the exchange resonance softened upon cooling and exhibited a redshift, contrary to the conventional blue shift with increased exchange interaction at low temperatures. This anomalous behavior was successfully reproduced using a three-sublattice LLG model incorporating a temperature-dependent anisotropic Yb-Fe exchange interaction. We trace this phenomenon to a modification of Yb-Fe exchange anisotropy, driven by the interplay of the Fe exchange field and Yb CEF excitations.

7.2 Outlook

The results presented in this thesis point towards several exciting directions for future research. First, the present study demonstrates that a combination of high Gilbert damping and suitable sublattice properties is essential for the experimental realization of the field-derivative torque. This provides a clear guideline for exploring other rare-earth garnets and multi-sublattice magnetic systems in which the relativistic contribution may be even more pronounced. Second, the population and coherence times of the exchange modes obtained in this work highlight the potential of rare-earth ferrimagnets for future THz spintronic and magnonic applications. The possibility of manipulating spin dynamics and precessional switching via relativistic torques may ultimately yield new design principles for ultrafast memory and information-processing technologies operating at terahertz frequencies.

Another promising direction arises from the strong coupling between CEF excitations and exchange dynamics observed in this thesis. The results suggest that magnetic anisotropy and exchange resonances can be dynamically controlled through resonant THz excitation of CEF states. More broadly, this work points towards a generalized material design strategy in which the THz spin dynamics of rare-earth iron garnets can be engineered by carefully selecting and tuning the rare-earth sublattice. Rare-earth ions with non-zero orbital angular momentum, such as Yb^{3+} , Ho^{3+} , Er^{3+} , and Tm^{3+} , can introduce CEF-mediated hybridization that substantially modifies the exchange-mode spectrum, in contrast to Y^{3+} and Gd^{3+} , which lack low-energy CEF excitations. Furthermore, the strength of hybridization, the resonance frequencies, and the temperature of spectral weight transfer can all be systematically tuned by the choice and concentration of the rare-earth ions. These findings open a pathway for tailoring exchange-controlled ultrafast magnetization dynamics and establish rare-earth iron garnets as a promising platform for next-generation THz spintronic and magnonic devices.

References

- [1] S. Parchenko. *Laser-induced Spin Dynamics in Multisublattice Ferrimagnetic Dielectrics*. PhD thesis, University of Bialystok, 2016.
- [2] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot. Ultrafast spin dynamics in ferromagnetic nickel. *Phys. Rev. Lett.*, 76(22):4250–4253, 1996.
- [3] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber. Coherent terahertz control of antiferromagnetic spin waves. *Nat. Photonics*, 5:31–34, 2011.
- [4] T. Kampfrath, M. Battiato, P. Maldonado, G. Eilers, J. Nötzold, S. Mährlein, V. Zbarsky, F. Freimuth, Y. Mokrousov, S. Blügel, M. Wolf, I. Radu, P. M. Oppeneer, and M. Münzenberg. Terahertz spin current pulses controlled by magnetic heterostructures. *Nat. Nanotechnol.*, 8(4):256–260, 2013.
- [5] J. Li, C.-J. Yang, R. Mondal, C. Tzschaschel, and S. Pal. A perspective on nonlinearities in coherent magnetization dynamics. *Appl. Phys. Lett.*, 120(5):050501, 2022.
- [6] J. Walowski and M. Münzenberg. Perspective: Ultrafast magnetism and thz spintronics. *J. Appl. Phys.*, 120(14):140901, 2016.
- [7] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, and Th. Rasing. Laser-induced ultrafast spin reorientation in the antiferromagnet TmFeO_3 . *Nature*, 429(6994):850–853, 2004.
- [8] B. Koopmans, J. J. M. Ruigrok, F. Dalla Longa, and W. J. M. de Jonge. Unifying ultrafast magnetization dynamics. *Phys. Rev. Lett.*, 95(26):267207, 2005.
- [9] C.-H. Lambert, S. Mangin, B. S. D. Ch. S. Varaprasad, Y. K. Takahashi, M. Hehn, M. Cinchetti, G. Malinowski, K. Hono, Y. Fainman, and M. Aeschli-mann. All-optical control of ferromagnetic thin films and nanostructures. *Science*, 345(6202):1337–1340, 2014.
- [10] T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. J. Heyderman, F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. A. Ivanov, A. M. Kalashnikova, K. Vahaplar, J. Mentink, A. Kirilyuk, Th. Rasing, and A. V. Kimel. Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet. *Nat. Commun.*, 3:666, 2012.

-
- [11] M. Battiato, K. Carva, and P. M. Oppeneer. Superdiffusive spin transport as a mechanism of ultrafast demagnetization. *Phys. Rev. Lett.*, 105(2):027203, 2010.
- [12] F. Willems, C. von Korff Schmising, C. Strüber, D. Schick, D. W. Engel, J. K. Dewhurst, P. Elliott, S. Sharma, and S. Eisebitt. Optical inter-site spin transfer probed by energy and spin-resolved transient absorption spectroscopy. *Nat. Commun.*, 11(1):871, 2020.
- [13] M. A. W. Schoen, D. Thonig, M. L. Schneider, T. J. Silva, H. T. Nembach, O. Eriksson, O. Karis, and J. M. Shaw. Ultra-low magnetic damping of a metallic ferromagnet. *Nat. Phys.*, 12(9):839–842, 2016.
- [14] H. Chang, P. Li, W. Zhang, T. Liu, A. Hoffmann, L. Deng, and M. Wu. Nanometer-thick yttrium iron garnet films with extremely low damping. *IEEE Magn. Lett.*, 5:1–4, 2014.
- [15] R. Salikhov, A. Alekhin, T. Parpiiev, T. Pezeril, D. Makarov, R. Abrudan, R. Meckenstock, F. Radu, M. Farle, H. Zabel, et al. Gilbert damping in NiFeGd compounds: Ferromagnetic resonance versus time-resolved spectroscopy. *Phys. Rev. B*, 99(10):104412, 2019.
- [16] S. K. Kim, G. S. D. Beach, K.-J. Lee, T. Ono, T. Rasing, and H. Yang. Ferrimagnetic spintronics. *Nat. Mater.*, 21(1):24–34, 2022.
- [17] N. Bhattacharjee, A. A. Sapozhnik, S. Y. Bodnar, V. Y. Grigorev, S. Y. Agustsson, J. Cao, D. Dominko, M. Obergfell, O. Gomonay, J. Sinova, et al. Néel spin-orbit torque driven antiferromagnetic resonance in Mn₂Au probed by time-domain thz spectroscopy. *Phys. Rev. Lett.*, 120(23):237201, 2018.
- [18] I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H. A. Dürr, T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, and A. V. Kimel. Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins. *Nature*, 472(7342):205–208, 2011.
- [19] C. E. Graves, A. H. Reid, T. Wang, B. Wu, S. de Jong, K. Vahaplar, I. Radu, D. P. Bernstein, M. Messerschmidt, L. Müller, R. Coffee, M. Bionta, S. W. Epp, R. Hartmann, N. Kimmel, G. Hauser, A. Hartmann, P. Holl, H. Gorke, J. H. Mentink, A. Tsukamoto, A. Fognini, J. J. Turner, W. F. Schlotter, D. Rolles, H. Soltau, L. Strüder, Y. Acremann, A. V. Kimel, A. Kirilyuk, Th. Rasing, J. Stöhr, A. O. Scherz, and H. A. Dürr. Nanoscale spin reversal by non-local angular momentum transfer following ultrafast laser excitation in ferrimagnetic GdFeCo. *Nat. Mater.*, 12(4):293–298, 2013.

-
- [20] T. G. H. Blank, K. A. Grishunin, E. A. Mashkovich, M. V. Logunov, A. K. Zvezdin, and A. V. Kimel. Thz-scale field-induced spin dynamics in ferrimagnetic iron garnets. *Phys. Rev. Lett.*, 127(3):037203, 2021.
- [21] M. C. Hickey and J. S. Moodera. Origin of intrinsic gilbert damping. *Phys. Rev. Lett.*, 102(13):137601, 2009.
- [22] R. Mondal, M. Berritta, and P. M. Oppeneer. Relativistic theory of spin relaxation mechanisms in the Landau-Lifshitz-Gilbert equation of spin dynamics. *Phys. Rev. B*, 94(14):144419, 2016.
- [23] D. Thonig and J. Henk. Gilbert damping tensor within the breathing fermi surface model: anisotropy and non-locality. *New J. Phys.*, 16(1):013032, 2014.
- [24] V. Kamberský. On the landau-lifshitz relaxation in ferromagnetic metals. *Can. J. Phys.*, 48(24):2906–2911, 1970.
- [25] B. Nagyfalusi, L. Szunyogh, and K. Palotás. Real-space nonlocal gilbert damping from exchange torque correlation applied to bulk ferromagnets and their surfaces. *Phys. Rev. B*, 109(9):094417, 2024.
- [26] E. Barati, M. Cinal, D. M. Edwards, and A. Umerski. Gilbert damping in magnetic layered systems. *Phys. Rev. B*, 90(1):014420, 2014.
- [27] J. Kuneš and V. Kamberský. First-principles investigation of the damping of fast magnetization precession in ferromagnetic 3d metals. *Phys. Rev. B*, 65(21):212411, 2002.
- [28] A. Sakuma. Theoretical investigation on the relationship between the torque correlation and spin correlation models for the gilbert damping constant. *J. Appl. Phys.*, 117(1):013912, 2015.
- [29] A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and Th. Rasing. Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses. *Nature*, 435(7042):655–657, 2005.
- [30] A. Kirilyuk, A. V. Kimel, and Th. Rasing. Ultrafast optical manipulation of magnetic order. *Rev. Mod. Phys.*, 82(3):2731–2784, 2010.
- [31] L. Q. Shen, L. F. Zhou, J. Y. Shi, M. Tang, Z. Zheng, D. Wu, S. M. Zhou, L. Y. Chen, and H. B. Zhao. Dominant role of inverse cotton-mouton effect in ultrafast stimulation of magnetization precession in undoped yttrium iron garnet films by 400-nm laser pulses. *Phys. Rev. B*, 97:224430, 2018.

-
- [32] J. Li, C.-J. Yang, R. Mondal, C. Tzschaschel, and S. Pal. A perspective on nonlinearities in coherent magnetization dynamics. *Appl. Phys. Lett.*, 120(5):050501, 2022.
 - [33] L. D. Landau and E. M. Lifshitz. On the theory of the dispersion of magnetic permeability in ferromagnetic bodies. *Phys. Z. Sowjetunion*, 8:153–169, 1935.
 - [34] L. D. Landau and E. M. Lifshitz. *Quantum Mechanics: Non-Relativistic Theory*, volume 3. Butterworth-Heinemann, Oxford, 3 edition, 1981.
 - [35] T. L. Gilbert and J. M. Kelly. Anomalous rotational damping in ferromagnetic sheets. In *Proceedings of the American Institute of Electrical Engineers*, pages 253–263, New York, October 1955.
 - [36] Z. Zhang, F. Y. Gao, J. B. Curtis, Z.-J. Liu, Y.-C. Chien, A. von Hoegen, M. T. Wong, T. Kurihara, T. Suemoto, P. Narang, et al. Terahertz field-induced nonlinear coupling of two magnon modes in an antiferromagnet. *Nat. Phys.*, 20(5):801–807, 2024.
 - [37] J. Lu, X. Li, H. Y. Hwang, B. K. Ofori-Okai, T. Kurihara, T. Suemoto, and K. A. Nelson. Coherent two-dimensional THz magnetic resonance spectroscopy of collective spin waves. *Phys. Rev. Lett.*, 118(20):207204, 2017.
 - [38] T. G. H. Blank, E. A. Mashkovich, K. A. Grishunin, C. F. Schippers, M. V. Logunov, B. Koopmans, A. K. Zvezdin, and A. V. Kimel. Effective rectification of terahertz electromagnetic fields in a ferrimagnetic iron garnet. *Phys. Rev. B*, 108(9):094439, 2023.
 - [39] D. Böttcher and J. Henk. Significance of nutation in magnetization dynamics of nanostructures. *Phys. Rev. B*, 86(2):020404, 2012.
 - [40] R. Mondal, M. Berritta, and P. M. Oppeneer. Signatures of relativistic spin-light coupling in magneto-optical pump-probe experiments. *J. Phys.: Condens. Matter*, 29(19):194002, 2017.
 - [41] R. Mondal, M. Berritta, and P. M. Oppeneer. Unified theory of magnetization dynamics with relativistic and nonrelativistic spin torques. *Phys. Rev. B*, 98(21):214429, 2018.
 - [42] K. Neeraj, M. Pancaldi, V. Scalera, S. Perna, M. d’Aquino, C. Serpico, and S. Bonetti. Magnetization switching in the inertial regime. *Phys. Rev. B*, 105(5):054415, 2022.
 - [43] V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, and S. Bonetti. Inertial spin dynamics in epitaxial cobalt films. *Phys. Rev. Lett.*, 129(23):237201, 2022.

-
- [44] R. Mondal, A. Donges, U. Ritzmann, P. M. Oppeneer, and U. Nowak. Terahertz spin dynamics driven by a field-derivative torque. *Phys. Rev. B*, 100(6):060409(R), 2019.
- [45] S. Parchenko, A. Stupakiewicz, I. Yoshimine, T. Satoh, and A. Maziewski. Wide frequencies range of spin excitations in a rare-earth bi-doped iron garnet with a giant faraday rotation. *Appl. Phys. Lett.*, 103(17):172402, 2013.
- [46] T. Satoh, Y. Terui, R. Moriya, B. A. Ivanov, K. Ando, E. Saitoh, T. Shimura, and K. Kuroda. Directional control of spin-wave emission by spatially shaped light. *Nat. Photonics*, 6(10):662–666, 2012.
- [47] S. Parchenko, M. Tekielak, I. Yoshimine, T. Satoh, A. Maziewski, and A. Stupakiewicz. Magnetization reversal and magnetic domain structures in Gd-Yb-BIG crystals. *IEEE Trans. Magn.*, 50(11):1–4, 2014.
- [48] A. Dutta, P. Mukherjee, S. P. Sarangi, S. Bhattacharjee, S. Pal, and R. Mondal. Role of material-dependent properties in THz field-derivative-torque-induced nonlinear magnetization dynamics. *Phys. Rev. Mater.*, 8(11):114404, 2024.
- [49] A. Dutta, C. Tzschaschel, D. Priyadarshi, K. Mikuni, T. Satoh, R. Mondal, and S. Pal. Evidence of relativistic field-derivative torque in nonlinear THz response of magnetization dynamics. *Adv. Funct. Mater.*, 35(35):2414582, 2025.
- [50] V. Peçanha-Antonio, D. Prabhakaran, C. Balz, A. Krajewska, and A. T. Boothroyd. Model for coupled $4f$ – $3d$ magnetic spectra: A neutron scattering study of the Yb-Fe hybridization in $\text{yb}_3\text{fe}_5\text{o}_{12}$. *Phys. Rev. B*, 105(10):104422, 2022.
- [51] A. J. Sievers III and M. Tinkham. Far infrared spectra of rare-earth iron garnets. *Phys. Rev.*, 129(5):1995–2004, 1963.
- [52] S. Blundell. *Magnetism in Condensed Matter*. Oxford University Press, Oxford, 2001.
- [53] S. Baierl, J. H. Mentink, M. Hohenleutner, L. Braun, T.-M. Do, C. Lange, A. Sell, M. Fiebig, G. Woltersdorf, T. Kampfrath, and R. Huber. Terahertz-driven nonlinear spin response of antiferromagnetic nickel oxide. *Phys. Rev. Lett.*, 117(19):197201, 2016.
- [54] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak. Antiferromagnetic spintronics. *Rev. Mod. Phys.*, 90(1):015005, 2018.
- [55] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich. Antiferromagnetic spintronics. *Nat. Nanotechnol.*, 11(3):231–241, 2016.

-
- [56] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and Th. Rasing. All-optical magnetic recording with circularly polarized light. *Phys. Rev. Lett.*, 99(4):047601, 2007.
- [57] E. A. Mashkovich, K. A. Grishunin, A. K. Zvezdin, T. G. H. Blank, A. G. Zavyalov, P. H. M. van Loosdrecht, A. M. Kalashnikova, and A. V. Kimel. Terahertz-driven magnetization dynamics of bismuth-substituted yttrium iron-gallium garnet thin film near a compensation point. *Phys. Rev. B*, 106(18):184425, 2022.
- [58] H. Bai, H.-A. Zhou, W. Li, T. Xu, L. Wang, P. Gargiani, M. Valvidares, and W. Jiang. 4f electron-mediated compensated magnetism in rare-earth-substituted iron garnet films with perpendicular magnetic anisotropy. *Phys. Rev. Appl.*, 23(4):044062, 2025.
- [59] K.-J. Kim, S. K. Kim, Y. Hirata, S.-H. Oh, T. Tono, D.-H. Kim, T. Okuno, W. S. Ham, S. Kim, G. Go, Y. Tserkovnyak, A. Tsukamoto, T. Moriyama, K.-J. Lee, and T. Ono. Fast domain wall motion in the vicinity of the angular momentum compensation temperature of ferrimagnets. *Nat. Mater.*, 16(12):1187–1192, 2017.
- [60] J. H. Mentink, J. Hellsvik, D. V. Afanasiev, B. A. Ivanov, A. Kirilyuk, A. V. Kimel, O. Eriksson, M. I. Katsnelson, and Th. Rasing. Ultrafast spin dynamics in multisub-lattice magnets. *Phys. Rev. Lett.*, 108(5):057202, 2012.
- [61] E. Haltz, J. Sampaio, S. Krishnia, L. Berges, R. Weil, A. Mougin, and A. Thiaville. Quantitative analysis of spin wave dynamics in ferrimagnets across compensation points. *Phys. Rev. B*, 105(10):104414, 2022.
- [62] C. D. Stanciu, A. V. Kimel, F. Hansteen, A. Tsukamoto, A. Itoh, A. Kirilyuk, and Th. Rasing. Ultrafast spin dynamics across compensation points in ferrimagnetic GdFeCo: The role of angular momentum compensation. *Phys. Rev. B*, 73(22):220402, 2006.
- [63] O. Gomonay, V. Baltz, A. Brataas, and Y. Tserkovnyak. Antiferromagnetic spin textures and dynamics. *Nat. Phys.*, 14(3):213–216, 2018.
- [64] A. Donges, S. Khmelevskiy, A. Deak, R.-M. Abrudan, D. Schmitz, I. Radu, F. Radu, L. Szunyogh, and U. Nowak. Magnetization compensation and spin reorientation transition in ferrimagnetic dyco_5 : Multiscale modeling and element-specific measurements. *Phys. Rev. B*, 96(2):024412, 2017.
- [65] K. Srinivasan, A. J. Grutter, T. E. Gage, P. Quarterman, C. J. Kinane, A. J. Caruana, G. Yu, J. Garcia-Barriocanal, S. Langridge, and B. J. H. Stadler. Interfacial magnetic characteristics of nearly compensated gadolinium iron garnet. *Phys. Rev. Mater.*, 8(4):044403, 2024.

-
- [66] M. Deb, P. Molho, B. Barbara, and J.-Y. Bigot. Controlling laser-induced magnetization reversal dynamics in a rare-earth iron garnet across the magnetization compensation point. *Phys. Rev. B*, 97(13):134419, 2018.
- [67] B. D. Cullity and C. D. Graham. *Introduction to Magnetic Materials*. John Wiley & Sons, Hoboken, NJ, 2 edition, 2011.
- [68] S. M. Rezende. *Fundamentals of Magnonics*, volume 969. Springer, Cham, 2020.
- [69] U. Nowak. Thermodynamics and statistical mechanics of classical spin systems. In H. Kronmüller and S. Parkin, editors, *Handbook of Magnetism and Advanced Magnetic Materials*. John Wiley & Sons, Chichester, 2007.
- [70] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, and R. W. Chantrell. Atomistic spin model simulations of magnetic nanomaterials. *J. Phys.: Condens. Matter*, 26(10):103202, 2014.
- [71] B. Skubic, J. Hellsvik, L. Nordström, and O. Eriksson. A method for atomistic spin dynamics simulations: Implementation and examples. *J. Phys.: Condens. Matter*, 20(31):315203, 2008.
- [72] A. Brataas, Y. Tserkovnyak, and G. E. W. Bauer. Scattering theory of gilbert damping. *Phys. Rev. Lett.*, 101(3):037207, 2008.
- [73] M.-C. Ciornei, J. M. Rubí, and J.-E. Wegrowe. Magnetization dynamics in the inertial regime: Nutation predicted at short time scales. *Phys. Rev. B*, 83(2):020410, 2011.
- [74] R. Mondal, L. Rózsa, M. Farle, P. M. Oppeneer, U. Nowak, and M. Cherkasskii. Inertial effects in ultrafast spin dynamics. *J. Magn. Magn. Mater.*, 579:170830, 2023.
- [75] K. Neeraj, N. Awari, S. Kovalev, D. Polley, N. Zhou Hagström, S. S. P. K. Arekapudi, A. Semisalova, K. Lenz, B. Green, J.-C. Deinert, I. Ilyakov, M. Chen, M. Bawatna, V. Scalera, M. d’Aquino, C. Serpico, O. Hellwig, J.-E. Wegrowe, M. Gensch, and S. Bonetti. Inertial spin dynamics in ferromagnets. *Nat. Phys.*, 17(2):245–251, 2021.
- [76] Z. Li and S. Zhang. Magnetization dynamics with a spin-transfer torque. *Phys. Rev. B*, 68(2):024404, 2003.
- [77] J. C. Slonczewski. Current-driven excitation of magnetic multilayers. *J. Magn. Magn. Mater.*, 159:L1–L7, 1996.
- [78] L. Berger. Emission of spin waves by a magnetic multilayer traversed by a current. *Phys. Rev. B*, 54(13):9353–9358, 1996.
- [79] D. C. Ralph and M. D. Stiles. Spin transfer torques. *J. Magn. Magn. Mater.*, 320(7):1190–1216, 2008.

-
- [80] Q. Shao, P. Li, L. Liu, H. Yang, S. Fukami, A. Razavi, H. Wu, K. Wang, F. Freimuth, Y. Mokrousov, M. D. Stiles, S. Emori, A. Hoffmann, J. Åkerman, K. Roy, J.-P. Wang, S.-H. Yang, K. Garello, and W. Zhang. Roadmap of spin-orbit torques. *IEEE Trans. Magn.*, 57(7):1–39, 2021.
- [81] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella. Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic systems. *Rev. Mod. Phys.*, 91(3):035004, 2019.
- [82] P. Gambardella and I. M. Miron. Current-induced spin–orbit torques. *Phil. Trans. R. Soc. A*, 369(1948):3175–3197, 2011.
- [83] G.-M. Choi, J. H. Oh, D.-K. Lee, S.-W. Lee, K. W. Kim, M. Lim, B.-C. Min, K.-J. Lee, and H.-W. Lee. Optical spin-orbit torque in heavy metal-ferromagnet heterostructures. *Nat. Commun.*, 11(1):1482, 2020.
- [84] R. Mondal, A. Donges, and U. Nowak. Terahertz spin dynamics driven by an optical spin-orbit torque. *Phys. Rev. Research*, 3(2):023116, 2021.
- [85] F. Hansteen, A. V. Kimel, A. Kirilyuk, and Th. Rasing. Nonthermal ultrafast optical control of the magnetization in garnet films. *Phys. Rev. B*, 73(1):014421, 2006.
- [86] P. S. Pershan, J. P. van der Ziel, and L. D. Malmstrom. Theoretical discussion of the inverse faraday effect, raman scattering, and related phenomena. *Phys. Rev.*, 143(3):574–583, 1966.
- [87] J. P. van der Ziel, P. S. Pershan, and L. D. Malmstrom. Optically-induced magnetization resulting from the inverse faraday effect. *Phys. Rev. Lett.*, 15(5):190–193, 1965.
- [88] Y. R. Shen and N. Bloembergen. Interaction between light waves and spin waves. *Phys. Rev.*, 143(2):372–381, 1966.
- [89] R Gómez-Abal, O Ney, K Satitkovitchai, and W Hübner. All-optical subpicosecond magnetic switching in nio (001). *Phys. Rev. Lett.*, 92(22):227402, 2004.
- [90] Y.-S. Lee. *Principles of Terahertz Science and Technology*. Springer, New York, 2009.
- [91] M. Kozina, M. Fechner, P. Marsik, T. van Driel, J. M. Glowina, C. Bernhard, M. Radovic, D. Zhu, S. Bonetti, U. Staub, et al. Terahertz-driven phonon upconversion in SrTiO₃. *Nat. Phys.*, 15(4):387–392, 2019.
- [92] H. Han, B. Li, S. Volz, and Y. A. Kosevich. Ultracompact interference phonon nanocapacitor for storage and lasing of coherent terahertz lattice waves. *Phys. Rev. Lett.*, 114(14):145501, 2015.

-
- [93] S. Sheng, M. Abdo, S. Rolf-Pissarczyk, K. Lichtenberg, S. Baumann, J. A. J. Burgess, L. Malavolti, and S. Loth. Terahertz spectroscopy of collective charge density wave dynamics at the atomic scale. *Nat. Phys.*, 20(10):1603–1608, 2024.
- [94] C.-J. Yang, J. Li, M. Fiebig, and S. Pal. Terahertz control of many-body dynamics in quantum materials. *Nat. Rev. Mater.*, 8(8):518–532, 2023.
- [95] X.-C. Zhang, X.-F. Ma, Y. Jin, T.-M. Lu, E. P. Boden, P. D. Phelps, K. R. Stewart, and C. P. Yakymyshyn. Terahertz optical rectification from a nonlinear organic crystal. *Appl. Phys. Lett.*, 61(26):3080–3082, 1992.
- [96] A. Rice, Y. Jin, X.-F. Ma, X.-C. Zhang, D. Bliss, J. Larkin, and M. Alexander. Terahertz optical rectification from $\langle 110 \rangle$ zinc-blende crystals. *Appl. Phys. Lett.*, 64(11):1324–1326, 1994.
- [97] A. Nahata, A. S. Weling, and T. F. Heinz. A wideband coherent terahertz spectroscopy system using optical rectification and electro-optic sampling. *Appl. Phys. Lett.*, 69(16):2321–2323, 1996.
- [98] R. W. Boyd. *Nonlinear Optics*. Academic Press, Cambridge, MA, 4 edition, 2020.
- [99] I. Wilke and S. Sengupta. Nonlinear optical techniques for terahertz pulse generation and detection: Optical rectification and electro-optic sampling. In *Terahertz Spectroscopy*. CRC Press, Boca Raton, FL, 2017.
- [100] P. C. M. Planken, H.-K. Nienhuys, H. J. Bakker, and T. Wenzelbach. Measurement and calculation of the orientation dependence of terahertz pulse detection in ZnTe. *J. Opt. Soc. Am. B*, 18(3):313–317, 2001.
- [101] O. Hatem. Peak emission of terahertz waves from (110)-oriented ZnTe by interacting phase-matched phonon resonances. *J. Opt. Soc. Am. B*, 36(4):1144–1149, 2019.
- [102] T. Elsaesser, K. Reimann, and M. Woerner. *Concepts and Applications of Nonlinear Terahertz Spectroscopy*. IOP Publishing, Bristol, UK, 2024.
- [103] A. Dutta, P. Shee, A. Halder, and S. Pal. 2D-THz spectroscopy: Exploring the nonlinear dynamics in quantum materials. *J. Phys.: Condens. Matter*, 37(20):203002, 2025.
- [104] K. A. Grishunin, V. R. Bilyk, E. D. Mishina, A. V. Kimel, and E. A. Mashkovich. Two-dimensional terahertz spectroscopy as a tool for revealing nonlinear interactions in media. *Rev. Sci. Instrum.*, 94(7):073005, 2023.

-
- [105] J. Raab, C. Lange, J. L. Boland, I. Laepple, M. Furthmeier, E. Dardanis, N. Dessmann, L. Li, E. H. Linfield, A. G. Davies, et al. Ultrafast two-dimensional field spectroscopy of terahertz intersubband saturable absorbers. *Opt. Express*, 27(3):2248–2257, 2019.
- [106] P. Mukherjee, A. Dutta, S. Bhattacharjee, S. Pal, and R. Mondal. Field-derivative torque induced magnetization reversal in ferrimagnetic $\text{Gd}_{3/2}\text{Yb}_{1/2}\text{BiFe}_5\text{O}_{12}$. *Phys. Rev. Mater.*, 9(10):104405, 2025.
- [107] P. Mukherjee and R. Mondal. Ultrafast switching of antiferromagnetic order by field-derivative torque. *Phys. Rev. B*, 113(5):054407, 2026.
- [108] O. I. Gorbatov, G. Johansson, A. Jakobsson, S. Mankovsky, H. Ebert, I. Di Marco, J. Minár, and C. Etz. Magnetic exchange interactions in yttrium iron garnet: A fully relativistic first-principles investigation. *Phys. Rev. B*, 104(17):174401, 2021.
- [109] L.-S. Xie, G.-X. Jin, L. He, G. E. W. Bauer, J. Barker, and K. Xia. First-principles study of exchange interactions of yttrium iron garnet. *Phys. Rev. B*, 95(1):014423, 2017.
- [110] L.-W. Wang, L.-S. Xie, P.-X. Xu, and K. Xia. First-principles study of magnon-phonon interactions in gadolinium iron garnet. *Phys. Rev. B*, 101(16):165137, 2020.
- [111] K. Mikuni, T. Hiraoka, T. Kuramoto, Y. Fujii, A. Koreeda, S. Parchenko, A. Stupakiewicz, and T. Satoh. Magnetic resonance frequency of two-sublattice ferrimagnet with magnetic compensation temperature. *J. Phys. Soc. Jpn.*, 94(11):111001, 2025.
- [112] S. Iida. Magnetostriction constants of rare earth iron garnets. *J. Phys. Soc. Jpn.*, 22(5):1201–1209, 1967.

Appendix A

Chapter 4 supplementary data

A.1 Birefringence of the material in THz frequency range

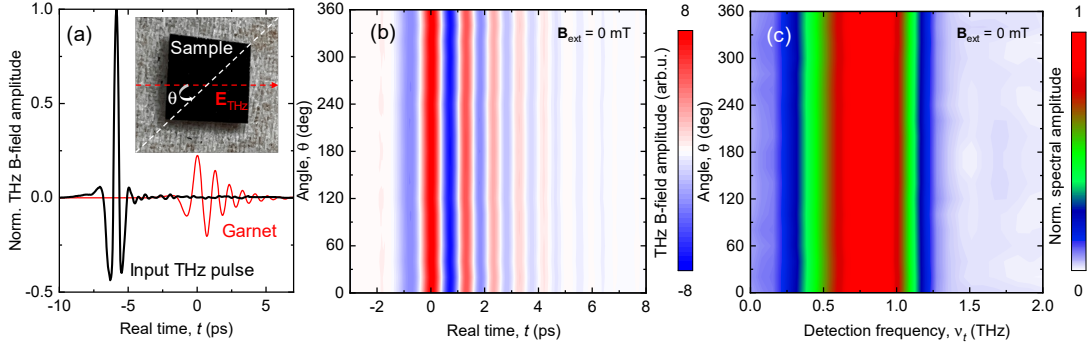


Figure A.1: (a) Time transients of input THz pulse and the transmitted FID signal. (inset) The corresponding sample geometry w.r.t. incident THz field. (b) The time profile and (c) the corresponding spectra of the transmitted FID signals as a function of the angle θ as shown in the inset of (a).

To rule out the possibility that crystal birefringence influences the orientation of the THz field with respect to the sample, we measured the THz transmittance as a function of the angle (θ) between the incident THz electric field polarization and the sample orientation. The sample was rotated through a full 360° in steps of 20° , while the incident THz polarization was kept fixed, as illustrated in the inset of Figure A.1a. All measurements were performed in the absence of an external static magnetic field.

Figures A.1b and c show that the transmitted THz waveforms and their corresponding frequency spectra remain essentially unchanged for all sample orientations, exhibiting no discernible signatures of crystal birefringence. In a birefringent medium, one would expect angle-dependent temporal delays and/or variations in the transmitted field amplitude. Similarly, in the frequency domain, birefringence would manifest as shifts in the spectral features and/or modulation of the spectral amplitude. Since no such effects are observed, we can confidently exclude crystal birefringence as a contributing factor in our measurements. This confirms that the observed excitation of the Kaplan-Kittel mode originates solely from the magnetic component of the incident THz field.

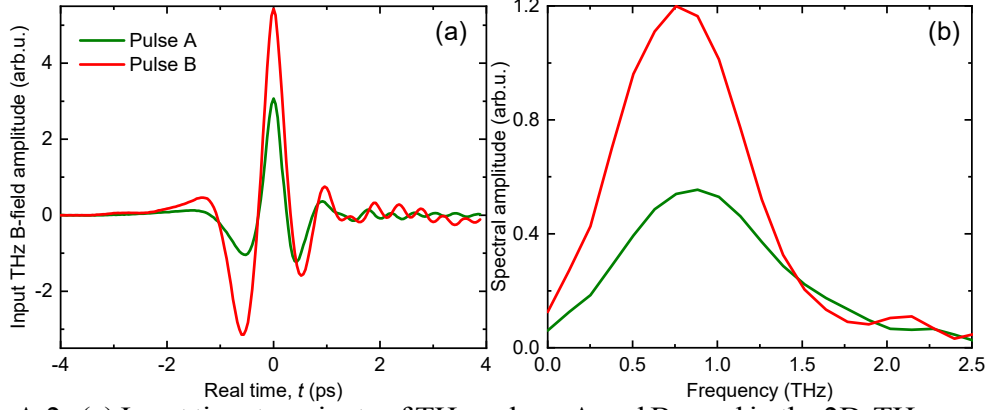


Figure A.2: (a) Input time transients of THz pulses, A and B, used in the 2D-THz experiment and (b) the corresponding THz spectra.

A.2 Input THz pulse and the corresponding spectra

The incident THz pulses are generated through optical rectification in 0.5-mm-thick ZnTe and GaP crystals, which serve as the sources of THz pulses A and B, respectively. The peak electric field of pulse B is approximately twice that of pulse A, while both pulses possess a comparable spectral bandwidth extending from 0.1 to 2.5 THz. The temporal profiles of the two incident THz pulses and their corresponding frequency spectra used in the two-dimensional THz spectroscopy measurements are presented in Figures A.2a and b, respectively.

A.3 Single THz pulse interactions

Figures A.3a, A.3b and A.3c show the 2D contour plot of the emitted individual responses $\mathbf{B}_B(t)$, $(\mathbf{B}_A(t, \tau))$ and $\mathbf{B}_{AB}(t, \tau)$ corresponding to the THz pulses B, A, and AB respectively. The two input THz pulses, namely $\mathbf{B}_A^{\text{in}}(t, \tau)$ and $\mathbf{B}_B^{\text{in}}(t)$ interact with the sublattice magnetizations of the garnet, resulting in the observed FID signals. The nonlinear signal is obtained by subtracting the individual field responses from both field response. The FID signals, resulting from the convolution of the Kaplan-Kittel exchange resonance and material absorption can be better observed by taking a horizontal cut in each of the 2D contour plots at a particular delay time, say at $\tau = 1.5$ ps as shown in Figure A.4.

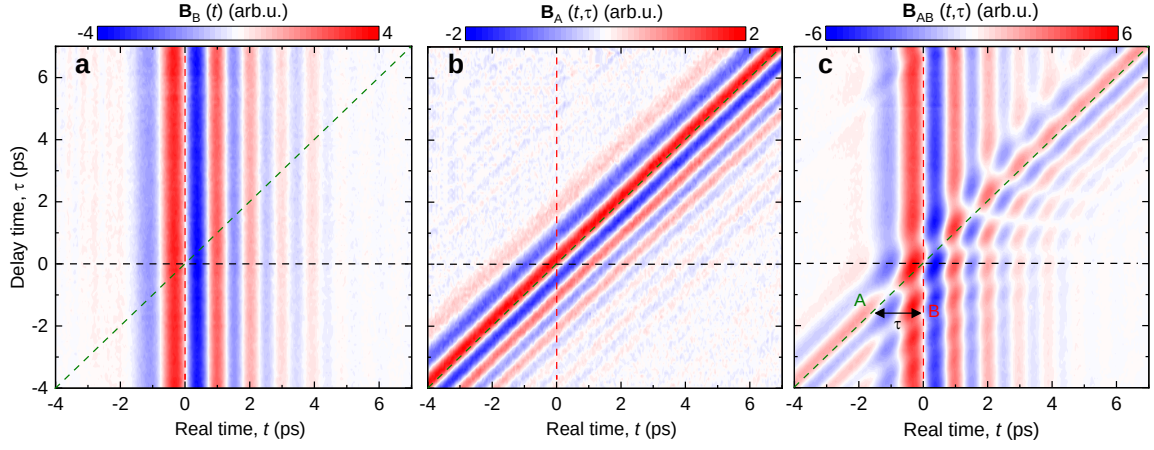


Figure A.3: Contour plots of transmitted THz responses from GdYbBIG of pulse B, A and AB, i.e., (a) $\mathbf{B}_B(t)$, $\mathbf{B}_A(t, \tau)$, and (c) $\mathbf{B}_{AB}(t, \tau)$ transmitted individually through the sample. The THz electric fields are plotted as a function of both the real time t and the delay time τ between the pulses. The green and red dashed lines indicate the position of the driving fields $\mathbf{B}_A$ and $\mathbf{B}_B$, respectively, while the black dashed line indicates zero delay time.

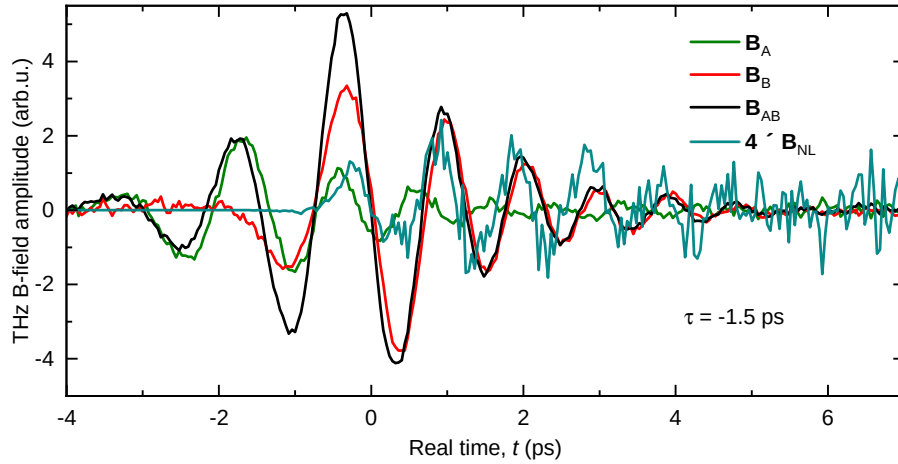


Figure A.4: An exemplary of the time transients corresponding to $\mathbf{B}_A$, $\mathbf{B}_B$, $\mathbf{B}_{AB}$, and $\mathbf{B}_{NL}$ at a delay time of $\tau = -1.5$ ps.

A.4 Inhomogeneous broadening

Among the four nonlinear features observed in the two-dimensional THz spectrum, the B-pump-A-probe signal exhibits a relatively larger linewidth compared to the other nonlinear peaks, indicating the presence of inhomogeneous broadening. In our experimental configuration, the peak electric field of THz pulse B is approximately twice that of THz pulse A. This asymmetry enables efficient access to the non-perturbative light-matter interaction

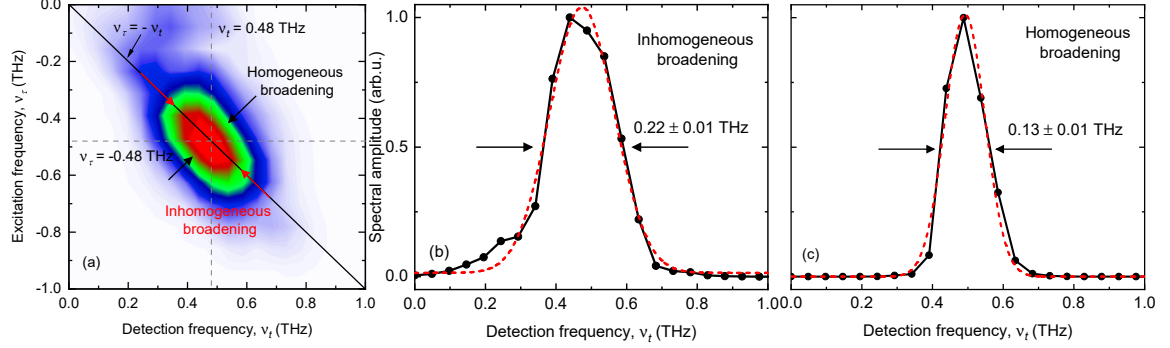


Figure A.5: (a) The B-pump-A-probe nonlinear signal. The black solid line along the diagonal represents the $\nu_e = -\nu_t$ line. The vertical and the horizontal gray dashed lines indicate the $\nu_t = 0.48$ THz and $\nu_e = -0.48$ THz lines, respectively. The (b) inhomogeneous and the (c) homogeneous profiles corresponding to the B-pump-A-probe nonlinear signal.

regime, thereby facilitating the generation and separation of the nonlinear responses. The comparatively stronger THz pulse B introduces a slight spatial variation in the effective magnetic field experienced by the sample, leading to an elongation of the B-pump-A-probe peak along the $(\nu_e = -\nu_t)$ direction, as shown in Figure A.5a. The corresponding inhomogeneous linewidth is estimated to be 0.22,THz (Figure A.5b), whereas the homogeneous linewidth is approximately 0.13,THz (Figure A.5c). The relatively small difference between these two linewidths indicates that homogeneous broadening remains the dominant contribution to the observed nonlinear response.

A.5 Field calibration of BNA-S

The amplitude of the input THz magnetic field is obtained from the relation $B_{\text{THz}} = E_{\text{THz}}/c$, where E_{THz} is the peak THz electric field and c is the velocity of light. The figure below shows the calibration data (from Terahertz Innovations) for the emitted THz field strength (kV/cm) versus the incident 800 nm pump fluence. We operated the BNA-S crystal in the linear regime, much below the laser-induced damage threshold. We have used a thick BNA-S crystal. In our experiments, we have pumped the crystal with 120 fs pulses at a fluence of 1.04 mJ/cm^2 at a repetition rate of 500 Hz. This corresponds to an electric field of 75 kV/cm, as per the datasheet, which is equivalent to a magnetic field of 0.25 mT.

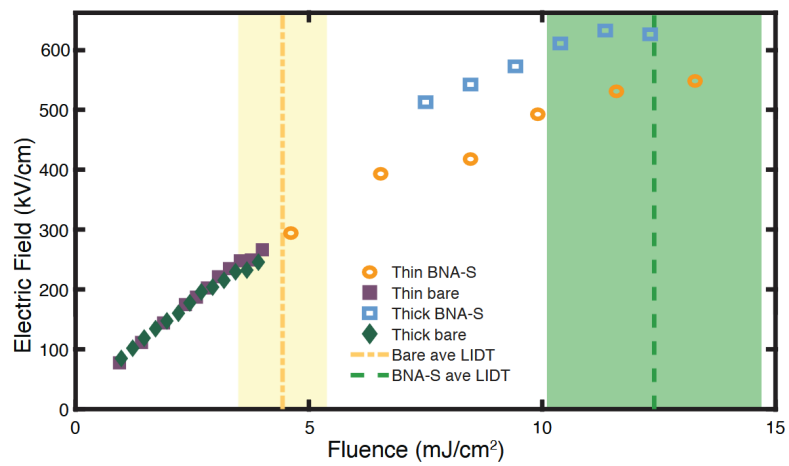


Figure A.6: Peak THz electric field as a function of 800 nm pump fluence at 500 Hz repetition rate. The figure is obtained from Terahertz Innovations (<https://www.terahertzinnovations.com/>).

Appendix B

Alignment procedure for the 2D-THz spectroscopy setup

This appendix summarizes the alignment procedure developed for the two-dimensional THz spectroscopy setup used throughout this work. The alignment is divided into two parts: (i) the primary THz beam path without the delay stage and (ii) the secondary THz beam path with the delay stage. Following the procedure described below significantly simplifies the alignment process and minimizes the spatial and temporal drifts during the measurements.

B.1 Alignment of the primary THz beam path (without delay stage)

Before starting the alignment, all optical components should be placed as close as possible to their final operating positions. This minimizes the shift in the temporal zero delay and facilitates locating the THz pulse during electro-optic detection.

B.1.1 THz generation and beam collimation

1. The THz beam generated through optical rectification should be centered on the first off-axis parabolic mirror immediately after the generation crystal. The mirror should be positioned exactly at its focal distance, which should coincide with the focal point of the focusing lens placed before the generation crystal. In our setup, the focal lengths of the lens and the first parabolic mirror were 150 mm and 156 mm, respectively.
2. The leakage beam immediately after the ZnTe crystal should exhibit a circular beam profile. A circular beam spot indicates proper collimation of the THz beam.

B.1.2 Verification of beam collimation

3. Place two irises to verify the beam collimation:

- one immediately after the first parabolic mirror,
- another immediately before the second parabolic mirror.

If the optical table holes cannot be used as references, place alignment cards with circular markings.

4. Align the beam at the first iris using the steering mirror before the generation crystal. Subsequently, align the beam at the second iris using the adjustment screws of the first parabolic mirror.

B.1.3 Alignment at the sample position

5. Position the second parabolic mirror such that its focal point coincides with the sample position. In our setup, the focal length of this mirror was 156 mm.
6. The beam should strike the focusing edge of the second parabolic mirror rather than its centre. This arrangement facilitates the later alignment of the second THz beam.
7. After the sample position, the diverging THz beam should be collected by the third parabolic mirror. At this stage, the beam should illuminate the opposite edge of the gold-plated mirror.
8. During both convergence toward and divergence from the sample, the beam profile should remain circular. Any noticeable ellipticity generally indicates astigmatism caused by improper alignment of the off-axis parabolic mirrors.

B.1.4 Detection beam path

9. Direct the reflected beam from the third parabolic mirror onto the plane mirror and subsequently into the hole parabolic mirror.
10. The beam should illuminate the focusing edge of the hole parabolic mirror without being clipped by its central aperture. Since the beam diameter generally increases along this path, sufficient clearance should always be maintained.
11. During the alignment, all mirrors should remain close to their final operating positions. This minimizes the shift of the temporal zero delay and simplifies locating the THz pulse during electro-optic detection.

B.2 Alignment of the secondary THz beam path (with delay stage)

After optimizing the primary THz beam path, the secondary beam line can be aligned.

B.2.1 Generation and collimation

1. The second THz beam should be generated using a focusing lens having a shorter focal length than that used for the primary beam. In our setup, the focal lengths were 125 mm and 150 mm for the secondary and primary beams, respectively, while both parabolic mirrors had focal lengths of 156 mm.
2. Position the second parabolic mirror at its focal distance and center the leakage beam on the mirror.
3. Since the two focusing lenses have different focal lengths, the centers of the first and second parabolic mirrors are separated by approximately 25 mm. Consequently, the two leakage beams also appear spatially separated.
4. Collimate the second THz beam following the same procedure described for the primary beam path.
5. Direct the second beam onto the opposite edge of the second parabolic mirror.
6. Verify that both THz beams focus at the same position on the sample. Properly collimated beams should produce nearly circular beam spots sharing a common focal plane. Typically, the primary beam appears slightly better focused than the secondary beam, which is acceptable.

B.2.2 Minimizing the beam separation

7. After the third parabolic mirror, both beams are reflected by the plane mirror. Initially, part of the beams may be clipped owing to their large spatial separation.
8. Reduce this separation by translating the stages supporting the first and second parabolic mirrors. Both stages should always be moved simultaneously in opposite directions. Avoid moving only one mirror.
9. Recheck the beam overlap at the sample position. Although the focal position may shift slightly, both beams should continue to share the same focal plane.

10. Further reduce the beam separation using the second axis of the XY translation stages supporting the first and fourth parabolic mirrors. Again, both stages should be translated simultaneously in opposite directions.
11. Slight ellipticity of the beam profile is acceptable. However, excessive ellipticity results in spatially inhomogeneous THz excitation and should therefore be avoided.

B.2.3 Final optimization

12. Correct any shift of the focal position using the steering mirrors before the generation crystals. Minor adjustments of the third and fourth parabolic mirrors and the plane mirror may also be required.
13. As a quick verification, place a white card immediately before the third parabolic mirror. If neither beam is clipped, both beams can be reflected by the plane mirror without significant losses.
14. Finally, optimize the overlap of the THz beams with the optical probe at the electro-optic crystal. If both THz beams are displaced together, adjust the hole parabolic mirror. If the THz and probe beams lie in different focal planes, translate the focusing lens in the probe beam path.
15. Adjustment of the first and second parabolic mirrors should be considered only as a last resort, since significant corrections at this stage disturb the beam overlap at the sample.
16. An additional degree of alignment freedom can be introduced by mounting the steering mirror before the second generation crystal on an XY translation stage. This allows fine spatial adjustment of the secondary beam without perturbing the downstream optical alignment.

Data Accessibility

The data shown in this thesis is archived at **\UTHD-Members\Arpita Dutta (PhD 2021-2026)\My Thesis** in the UTHD group's internal server. The folder is organized according to the Chapters of this thesis. All data supporting the findings of this thesis are available from the author upon reasonable request.

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Personal Statement

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- Carrying out low-temperature optical, THz, and magneto-optical measurements to investigate strongly correlated magnetic systems.
- Designing, assembling, and optimizing state-of-the-art time-resolved spectroscopy setups, including optical pump-THz probe and magneto-optical detection schemes.

Supervision and Mentoring

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Master's Project: Interplay between charge carrier dynamics and magnetism in a 2D Van-der Waals system $\text{Cr}_2\text{Ge}_2\text{Te}_6$.

- **Mr. Srijan Maity** (2025–Present)

Master's Project: Linear and non-linear THz magnetization dynamics in HoFeO_3 .

- **Mr. Swosti Prakash Sarangi** (2023–2025)

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- **Ms. Somasree Bhattacharjee** (June 2024–July 2024)

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Physics First Year Laboratory

Faculty: Dr. Ashis Kumar Nandy

August 2023–December 2023

Electromagnetic Theory

Faculty: Dr. Prolay Mal and Dr. Nishikanta Khandai

January 2024–August 2024

Refereed Journal Publications

Publications Related to Ph.D. Thesis

* Equal contribution

1. **A. Dutta***, P. Mukherjee*, R. Mondal, and S. Pal, *The interplay of crystal-field transitions and exchange spin dynamics in a ferrimagnet*, (Manuscript accepted in Advanced Physics Research).
2. **A. Dutta**, C. Tzschaschel, D. Priyadarshi, K. Mikuni, T. Satoh, R. Mondal, and S. Pal, *Evidence of relativistic field-derivative torque in nonlinear THz response of magnetization dynamics*, Advanced Functional Materials **35**, 2414582 (2025).
3. **A. Dutta**, P. Mukherjee, S. P. Sarangi, S. Bhattacharjee, S. Pal and R. Mondal, *Role of material-dependent properties in THz field-derivative-torque-induced nonlinear magnetization dynamics*, Physical Review Materials **8**, 114404 (2024).

Collaborative Publications and Review Articles

1. J. Li, **A. Dutta**, K. Saha, A. Szczerbakow, T. Story, M. Fiebig, and S. Pal, *Spectroscopic signatures of a Kohn anomaly in a topological crystalline insulator*, Physical Review B **114**, 225111 (2026).
2. K. Mondal, A. Halder, I. Mondal, **A. Dutta**, H. Bhatt, B. Mahata, S. Banerjee, H. N. Ghosh, S. P. Senanayak, and S. Pal, *Intensity-driven Mott-like transition in a 2D Ruddlesden-Popper perovskite*, (Manuscript accepted in Small).

3. P. Mukherjee, **A. Dutta**, S. Pal, and R. Mondal, *Field-derivative torque induced magnetization reversal in ferrimagnetic $Gd_{3/2}Yb_{1/2}BiFe_5O_{12}$* , Physical Review Materials **9**, 104405 (2025).
4. **A. Dutta**, P. Shee, A. Haldar, and S. Pal, *2D-THz spectroscopy: Exploring the nonlinear dynamics in quantum materials*, Journal of Physics: Condensed Matter **37**, 203002 (2025).
5. K. Jagadish, A. Godha, B. Pandit, Y. Jadhav, **A. Dutta**, J. Satapathy, H. Bhatt, B. Singh, S. K. Makineni, S. Pal and S. R. Rondiya, *Charge carrier dynamics in bandgap modulated covellite-CuS nanostructures*, Small **20**, 2405859 (2024).
6. A. Puthukkudi, S. Nath, P. Shee, **A. Dutta**, C. V. Rajput, S. Bommakanti, J. Mohapatra, M. Samal, S. Anwar, S. Pal, and B. P. Biswal, *Terahertz Conductivity of Free-Standing 3D Covalent Organic Framework Membranes Fabricated via Triple-Layer-Dual Interfacial Approach*, Advanced Materials **36**, 2312960 (2024).

Conference Presentations

1. *Poster:*
Evidence of relativistic field-derivative torque in nonlinear THz response of magnetization dynamics, Indian Materials Conclave and International Conference on Emerging Materials (MRSI-ICEM), NISER Bhubaneswar, India, 17-20 December 2025.
2. *Invited Talk:*
Exploring the ultrafast spin dynamics in ferrimagnets and antiferromagnets, An International Symposium on Ultrafast Spintronics and Magnonics (ISUSM), IIT-ISM Dhanbad, India, 2-3 February 2025.
3. *Contributory Talk:*
Exploring the ultrafast dynamics of quantum materials, School of Physical Science Day (SPS Day), NISER Bhubaneswar, India, 30-31 August 2024.
4. *Poster:*
Experimental observation of field derivative torque in nonlinear THz response of magnetization dynamics, Physics of Strongly Correlated Electron System (PSCES), IISER Bhopal, India, 8-10 April 2024.
5. *Poster:*
Microscopic picture of THz exchange nonlinearities in a ferrimagnetic insulator, Quantum Condensed Matter (QMAT), NISER Bhubaneswar, India, 27-30 November 2023.
6. *Poster:*
Observation of room temperature THz magnons in a ferrimagnetic garnet, Ultrafast Sciences (UFS), IISER Trivandrum, India, 3-5 November 2022.

Awards and Scholarships

- Best Poster Award in Indian Materials Conclave and International Conference on Emerging Materials (MRSI-ICEM) 2025
- Best Oral Presentation Award in School of Physical Science Day, National Institute of Science Education and Research (SPS-NISER) 2024
- Best Poster Award in Quantum Condensed Matter (QMAT) 2023
- Honorable Mention in Ultrafast Sciences (UFS) 2022
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