

Abstract

The exponential growth of data generation in the modern era has placed unprecedented demands on memory and computing technologies, driving an urgent need for storage devices that are faster, energy-efficient, and more scalable than existing charge-based technologies. Conventional magnetic memory, from hard disk drives to dynamic random-access memory, is fundamentally limited by thermodynamic and Joule-heating penalties associated with charge-current-driven magnetization switching. Spintronics, which exploits the electron's spin degree of freedom in addition to its charge, has progressively addressed these limitations through magnetic random-access memory (MRAM), spin-transfer torque (STT), spin-orbit torque (SOT), and voltage-controlled magnetic anisotropy (VCMA) devices. Yet even the most advanced of these technologies remains constrained by the need to drive charge through a magnetic layer, with switching speeds bounded in the nanosecond regime and energy costs in the picojoule-per-bit range. Surpassing these limits requires a completely different physical mechanism, one that can manipulate the magnetic exchange interaction itself. This thesis investigates coherent phonon excitation as such a mechanism: a non-dissipative, sub-picosecond, frequency-tunable route to engineering magnetic exchange interactions and spin-wave spectra, with direct implications for next-generation ultrafast spintronic memory and quantum information devices.

The central physical idea is that the magnetic exchange interaction between neighboring magnetic ions is not a fixed material constant but a sensitive function of local atomic geometry, bond angles, bond lengths, and orbital overlaps. A pulsed terahertz (THz) or mid-infrared (MIR) electric field tuned to a lattice resonance can coherently drive atoms along a specific phonon eigenvector on a sub-picosecond timescale, modifying the bond geometry, renormalizing the exchange interaction, and the spin-wave spectrum, all without heating the electronic system or driving any charge current. The symmetry of the excited phonon mode determines which exchange pathways are perturbed and in what pattern, making the excitation frequency a design variable for selectively engineering the spin Hamiltonian. This thesis tries to answer the following

questions: how exactly does a coherent lattice vibration renormalize the magnetic exchange interaction, and which orbital channels carry that renormalization? Can phonon mode symmetry be exploited as a design degree of freedom to selectively target specific exchange pathways? How do the renormalized exchange interactions feed back into the phonon spectrum, and can this feedback be experimentally observed and quantified? Can coherent phonon excitation change spin states without a charge current?

To answer these questions, this thesis applies a Wannier-orbital-based Green's function framework that resolves the exchange interaction into contributions from individual d -orbitals, each carrying FM or AFM character depending on its overlap geometry with the bridging ligand, and recalculates these contributions for phonon-perturbed atomic configurations. The renormalized exchange constants are used as input to linear spin-wave theory to obtain the full magnon dispersion, and are further connected to phonon frequency renormalization through the coupled spin-lattice Hamiltonian, enabling quantitative comparison with anomalous phonon shifts in temperature-dependent Raman and infrared spectroscopy experiments across the material systems.

We begin with the archetypal antiferromagnet NiO, where the relatively simple superexchange geometry allows us to establish the foundational methodology and uncover a key result that underpins all subsequent work: the exchange interaction varies quadratically, and not linearly, with atomic displacement, primarily through the $d_{3z^2-1} - p_z - d_{3z^2-1}$ orbital channel, in contrast to the widely used linear approximation. By resolving the spin-phonon coupling by both phonon mode and orbital channel, we determine the changes in phonon and magnon energy, group velocity, and phonon-magnon scattering amplitude. Building on this foundation, we extend the framework to the cubic fluoroperovskite KNiF₃, where the higher crystal symmetry provides a clean setting to demonstrate the mode-selectivity of coherent phonon excitation. We show that resonant phonon excitation renormalizes magnon energies across the full Brillouin zone, while selectively lifting the degeneracy of otherwise equivalent exchange interactions, a capability other photoexcitations are unable to provide. The renormalized exchange constants are further shown to quantitatively reproduce anomalous phonon shifts observed in infrared spectroscopy across the magnetic ordering temperature, validating the coupled spin-phonon Hamiltonian against experiment. We then turn to the antiferromagnetic oxide YCrO₃, a complex orbital system in which all five partially occupied Cr d orbitals simultaneously contribute to the exchange interaction through competing ferromagnetic and antiferromagnetic channels. Selective

narrow- and finite-bandwidth phonon excitation is shown to perturb the orbital overlaps and their relative contributions to FM and AFM interactions, lift exchange degeneracies, renormalize the exchange constants, and significantly alter the spin waves across the Brillouin zone. A central finding is that the symmetry of the excited phonon mode determines which exchange pathways are perturbed and in what spatial pattern, while finite-bandwidth multimode excitation accesses magnetic configurations unattainable by any single-mode perturbation, thereby directly establishing the theoretical basis for frequency-tunable, mode-selective spin control. The renormalized exchange constants further modify the interatomic force constants and phonon frequencies, again in quantitative agreement with spectroscopy measurements.

Finally, departing from spin-phonon coupling to the broader theme of nonlinear phonon dynamics, we investigate the conditions required for the spontaneous formation of phonon frequency combs in van der Waals solids. While phononic frequency combs have been demonstrated experimentally and theoretically in the kHz-MHz regime under nonlinear driving, their spontaneous formation in the GHz-THz regime remains rare. Our spectral energy density calculations using machine-learning-augmented molecular dynamics simulations provide evidence for frequency combs in the flat $A_g(5)$ phonon mode of CrGeTe_3 . However, the comb spacings are comparable to the phonon linewidth, making precise identification of the frequency comb challenging. From a comprehensive analysis of different modes of CrGeTe_3 , we identified two conditions to distinctively observe the combs beyond the experimental resolution: (i) an intramolecular or isolated mode with reasonably large third- or higher-order bond anharmonicity and (ii) limited phonon scattering channels. Our analysis of other van der Waals solids highlighted that these conditions are met for the nearly flat intramolecular E_{2g} and A_{2u} phonon modes of WSe_2 . Available Raman scattering data of the E_{2g} mode in the literature provide preliminary, if not conclusive, evidence of comb formation.

Collectively, the results of this thesis establish that coherent phonon excitation offers a physically compelling and theoretically validated pathway to spintronic switching at speeds below 1 ps, energy costs approaching the femtojoule-per-bit regime, and with a mode-selective design space for engineering spin Hamiltonians that is inaccessible to any charge-current-based technology. In addition to identifying design rules for spontaneous phononic frequency combs, this work provides a pathway toward phonon-engineered quantum materials.