Nonlinear coherent spin dynamics in antiferromagnets initiated by Tesla-class terahertz magnetic fields

Zhenya Zhang

A good understanding of spin dynamics is essential for the design and development of a variety of spintronic devices for information transport, processing, and recording. In contrast to ferromagnets, antiferromagnets have the advantage of an intrinsic high-speed spin precession at terahertz (THz) frequencies as a result of the robust antiferromagnetic exchange interaction. Thus, ultrafast control of antiferromagnetic spins has attracted considerable interest. Initially, the use of circularly polarized near-infrared pulses emerged as a viable method for coherent spin excitation because a magnetic field can be generated through the inverse Faraday effect. Subsequently, alternative methods were discovered. For instance, it has been shown that the exchange interaction can be modified by exciting charge-transfer electronic transitions or lattice vibrations. However, these methods often lead to significant heating, because they strongly excite either electrons or phonons. Since 2010, the use of THz pulses (usually in the range of $0.1–3\times10^{12}$ Hz) has emerged as a method for coherent spin excitation that does not involve a strong excitation of electrons or phonons, because these pulses have low photon energies. The feasibility of coherent spin excitation using THz electric fields has been demonstrated by resonantly exciting a crystal-field-split electronic transition within the ground-state doublet of a rare-earth ion in an orthoferrite. This method is limited to materials with crystal-field split transitions at THz frequencies as well as sufficiently low temperatures where a thermal population that hinders THz excitation is not created. The use of THz magnetic fields can be regarded as the most general method to coherently excite antiferromagnetic spins in various materials and temperature ranges. However, due to the relatively weak magnetic field of THz pulses propagating in free space, many experiments on nonlinear coherent spin dynamics in antiferromagnets can only investigate a restricted range of phenomena.

In this thesis, we present our investigations on the nonlinearities of the coherent spin dynamics in the rare-earth orthoferrites $HoFeO_3$ and $Sm_{0.7}Er_{0.3}FeO_3$ near room temperature using Tesla-class THz magnetic fields. Both $HoFeO_3$ and $Sm_{0.7}Er_{0.3}FeO_3$ are canted antiferromagnets with a small net magnetization. The spin dynamics in these antiferromagnets can be described in terms of their intrinsic modes: the quasi-antiferromagnetic (q-AFM) and the quasi-ferromagnetic (q-FM) modes. The nonlinearities of the q-AFM mode in $HoFeO_3$ and the q-FM mode in $Sm_{0.7}Er_{0.3}FeO_3$ were studied using the same experimental method: To generate a Tesla-class THz magnetic field in the sample, we designed a spiral-shaped microresonator and fabricated it on the sample surface by electron-beam lithography. When the microresonator is irradiated with a THz pulse, an oscillating current is induced that generates a strong multicycle THz magnetic near-field perpendicular to the sample surface. The coherent spin dynamics initiated by the THz magnetic near-field are detected by measuring the change in the ellipticity angle ($\Delta\eta$) of an 800-nm probe pulse due to the magneto-optic effect.

Firstly, in the case of $HoFeO_3$, we chose a crystal orientation where the net magnetization is parallel to the magnetic near-field to ensure that only the q-AFM mode is excited. The time-resolved magneto-optic signal $\Delta\eta$ showed an oscillation at about 0.58 THz, the frequency of the q-AFM mode. The following nonlinear phenomena were observed: (a) The upper and lower envelopes of the oscillation of $\Delta\eta$ become more asymmetric as the field strength is increased; apart from the fundamental peak at about 0.58 THz, the corresponding spectrum contains the second- and third-order harmonics, and their integrated peak areas obey a power law. (b) The q-AFM-mode frequency exhibits a redshift as the field strength is increased. We investigated the origins of these nonlinear phenomena by solving the Landau–Lifshitz–Gilbert (LLG) equation for a two-sublattice model. We found that, under strong excitation conditions, the q-AFM mode cannot be approximated as a harmonic oscillator. The observation of the third-order harmonic clearly verifies the anharmonicity of the potential. Due to the sinusoidal potential, the frequency of the q-AFM mode exhibits a redshift as the field strength is increased. From the value of the redshift observed in the experiment, we estimated a maximum spin deflection angle of about 34 degrees away from the equilibrium orientation. The asymmetric envelopes and the second-order harmonic are attributed to the nonlinearities of the magneto-optic effect related to the q-AFM mode and can be reproduced well by the numerical solution of the LLG equation and the wave equation for the probe pulse propagating through the sample. These results reveal the nonlinearities of q-AFM modes in antiferromagnets.

Secondly, in the case of $Sm_{0.7}Er_{0.3}FeO_3$, the magnetic near-field (with a spectral peak at about 0.46 THz) forms an angle with the net magnetization that lies between 0 and 90 degrees, depending on the sample temperature. Above 310 K, the time-resolved magneto-optic signal $\Delta\eta$ showed a superposition of the q-FM (<0.05 THz) and the q-AFM (~0.5 THz) modes. We found the following behaviors: (a) As the field strength is increased beyond a critical value, a long-lived offset emerges after the first half cycle of the q-FM mode, and the amplitude of this offset exhibits a steep dependence on the field strength. (b) The offset changes drastically with the temperature, which controls the magnetic potential of the q-FM mode (we can control the barrier height between the two potential minima in $Sm_{0.7}Er_{0.3}FeO_3$); the offset only appears when the potential barrier is small enough. These two features of the offset reveal that spin switching occurs in this system after sufficiently strong excitation, that is, the magnetic order parameter overcomes the barrier and reaches the second potential minimum. We derived and solved the sine-Gordon equation based on the LLG equation to reproduce the q-FM-mode dynamics including the spin-switching phenomenon. This sine-Gordon equation includes the effect of the Zeeman torque directly exerted by the magnetic near-field and that of coupling between the q-AFM and q-FM modes. To explain why the offset emerges after the first half q-FM-mode cycle, we analytically solved the sine-Gordon equation by replacing the expression for the potential modified by the fast-oscillating magnetic field with a "time-averaged" effective potential that varies with the envelope of the field. We found that the effective potential initially drives the magnetic order parameter away from the barrier, and when the potential modification vanishes, the magnetic order parameter starts to approach the barrier and overcomes it due to the inertia effect. These results clarify the nonlinearities of the q-FM mode in this antiferromagnet.

Finally, in this thesis, we summarize the nonlinear coherent dynamics of the q-AFM and q-FM modes in $HoFeO_3$ and $Sm_{0.7}Er_{0.3}FeO_3$, respectively. The observed frequency redshift and the third-order harmonic of the q-AFM mode reflect the anharmonic potential energy for the q-AFM mode. Regarding the q-FM mode, we observed an extremely nonlinear phenomenon, spin switching. The excitation process responsible for spin switching is an intriguing nonlinear process where a multicycle magnetic near-field modifies the potential for the q-FM mode. Similar processes can also occur in electronic systems, and this type of control through multicycle fields is also known as Floquet engineering. Our investigations show that THz magnetic near-fields are potentially useful for the investigation of coherent spin nonlinearities and novel methods for ultrafast manipulation of spin states.