Experimental observation of sub-THz magnon fluctuations in an antiferromagnet and spontaneous spin switching

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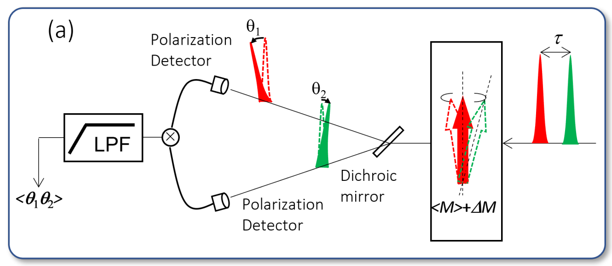
Many elementary excitations in solids such as phonons, excitons, plasmons and magnons exhibit eigenfrequencies in the THz region. In magnetically ordered spin systems such as ferro- and antiferromagnets, the spontaneous fluctuations of spins induced by quantum mechanical interactions and the thermal population play crucial roles in a wide range of fundamentally important phenomena ranging from magnetic phase transition, spin caloritronic effects and stochastic switching. However, they are mostly dismissed in conventional pump-probe measurements because averaging the probe signals through multiple repetitions tends to smear out their contributions due to their inherently random nature.

Recently, we have developed an experimental system that allows to resolve the THz spin fluctuation dynamics directly on ultrafast time scales with femtosecond resolution [Figure 1 (a)] [1]. Inspired by the emerging field of sub-cycle quantum optics [2], the central idea of the system relies on measuring the statistical correlation of the pulse-to-pulse polarization fluctuations contained in a pair of two-color optical probe pulses transmitted through the sample [3]. As the benchmark test sample, we used a custom synthesized crystal of the mixed orthoferrite, $Sm_{0.7}Er_{0.3}FeO_3$ [4]. It is a weak ferromagnet with a slightly canted antiferromagnetic order and exhibits weak net magnetization M. The natural resonance modes lie in the sub-THz region. The sample stoichiometry is tuned such that the second-order rotational type spin reorientation (SRT) occurs close to room temperature.



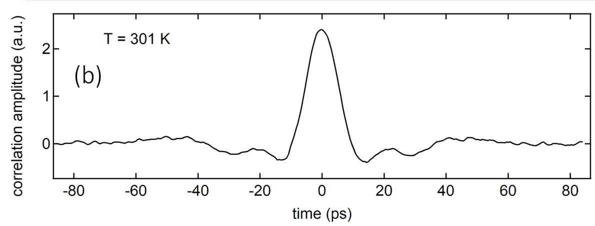


Fig. 1: (a) Schematic of the ultrafast spin noise experiment. LPF: low-pass filter. (b) Magnon autocorrelation waveform of $Sm_{0.7}Er_{0.3}FeO_3$ measured at 301 K.

Figure 1 (b) shows a typical autocorrelation trace measured at a temperature in the vicinity of the SRT. The waveform exhibits an oscillation and a finite coherence time reaching tens of picoseconds, which quantitatively matches the quasi-ferromagnetic mode magnon oscillation period and is significantly longer than the probe pulse duration (< 300 fs). The waveform drastically changes around the SRT, confirming that the detected signal originates from the incoherent fluctuations of magnetization. Especially, within the SRT temperature range we observed a significant divergence of the amplitude and coherence time of the waveform. Comparison with a large-scale atomistic simulation of spin dynamics ascribes this feature to a spontaneous switching of the weak magnetization between the two equilibrium states of the double-well magnetic anisotropy potential.

References:

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