Ultrafast magnetization reversal by excitation at the frequencies of optical phonons (in-depth report)

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Identifying an efficient pathway to change the order parameter via a subtle excitation of the coupled high-frequency mode is the ultimate goal of the field of ultrafast phase transitions [1,2]. This is an especially interesting research direction in magnetism, where the coupling between spin and lattice excitations is required for magnetization reversal [3]. Control of the crystal environment arguably represents the most universal mechanism to act on magnetization, as it is present in all materials regardless of their magnetic structure. One interesting approach to manipulate crystal lattice is via the anharmonic interaction of different phonon modes, which transfers the high-frequency excitation of an infrared-active mode into a rectified displacement along a coupled Raman-active coordinate [4,5].

To provide resonant excitation of the optical phonon modes, we use pulses from FELIX (Free Electron Lasers for Infrared eXperiments, Nijmegen, The Netherlands). Single pulses of IR/THz light with wavelength in the range of 10-50 μ m are typically used. The pulses of FELIX have been shown to be Fourier-transform limited [6], with their bandwidth experimentally tunable in the range of 0.2-2.0%, corresponding to the typical pulse width of 1-10 ps, depending on the wavelength range.

And thus we show how an ultrafast resonant excitation of the longitudinal optical phonon modes in magnetic garnet films switches magnetization into a peculiar quadrupolar magnetic domain pattern, unambiguously revealing the magneto-elastic mechanism of the switching [7]. In contrast, the excitation of strongly absorbing transverse phonon modes results in thermal demagnetization effect only. The mechanism appears to be very universal, and is shown to work in samples with very different crystallographic symmetry and magnetic properties, including weak ferromagnets and antiferromagnets [8], but also completely different systems such as ferroelectrics. Using single-shot time resolved microscopy, we demonstrate that the dynamics of the domain formation proceeds via a strongly inhomogeneous magnetic state and formation of magnon-polarons [9]. This we could arguable relate to the spin-wave instabilities that appear due to the very large amplitude of precessional magnetic motion in the switching process [10].

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