



SFB1242

Nichtgleichgewichtsdynamik kondensierter
Materie in der Zeitdomäne

UNIVERSITÄT
DUISBURG
ESSEN

Open-Minded

07.11.2023 / 10 Uhr c.t., Raum MG 272
Campus Duisburg

Non-thermal engineering of solid-state systems via THz driven collective modes

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The possibility of designing properties on-demand in solid-state systems using mode-selective electromagnetic drives has motivated a series of studies to investigate the interplay between different microscopic degrees of freedom. First experiments achieved this type of control through nonlinear coupling between a resonantly driven infrared active mode and a second mode which is interconnected with a macroscopic material property, so-called nonlinear phononics. However, so far the non-linear dynamics of the driven mode itself remained hidden.

We used time-resolved second harmonic generation (tr-SHG) to directly measure the large amplitude oscillations of a THz driven infrared-active mode in the ferroelectric material LiNbO_3 . The amplitude and phase sensitive detection of the lattice dynamics allowed us to reconstruct the lattice potential energy.

We then applied this scheme to the XPS_3 ($X=\text{Mn, Ni, Fe, Co}$) family of van der Waals antiferromagnets. This class of materials recently attracted a lot of attention due to their strongly coupled spin, lattice and electronic degrees of freedom. We use intense THz light pulses to resonantly drive the fundamental eigenmodes of the spin and lattice degrees of freedom, i.e., magnons and phonons, in FePS_3 directly. Our THz excitation launches spin and lattice dynamics, giving rise to coherent magnons and phonons, which we follow as a function of temperature by recording the polarization rotation (dichroism) and ellipticity (birefringence) of a transmitted 800 nm laser pulse. Besides these coherent dynamics, we also observe the emergence of a slow component close to the Neel temperature $T_N = 118 \text{ K}$. These slow dynamics are concomitant with the appearance of a long-lived circular dichroism (CD) are evidence for a finite out-of-plane magnetization inside the sample. These findings demonstrate how the magnetic ground state in 2D van der Waals magnets can be efficiently manipulated along non-thermal pathways using THz light.

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