Nonlinear phononics and phono-magnetism in iron oxides

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Controlling quantum phases of matter at fastest possible timescales is a major challenge in condensed matter physics. Recently, resonant femtosecond excitation of coherent optical phonons has become a powerful technique to control the properties of quantum materials [1]. In the last few years it was demonstrated that strong ultrafast driving of infrared active phonons in magnetic oxides can also provide a new route to control magnetic properties of materials by triggering coherent magnon oscillations [2] or even inducing a transient magnetic phase not accessible by adiabatic changes of ambient conditions [3]. Thus a new field of "phono-magnetism" has been opened (Fig. 1).

Here, we report the coherent phono-magnetism in a broad class of iron oxides: canted antiferromagnets such as orthoferrites and iron borate as well as ferrimagnetic magnetite. We perform our experiments in a pump-probe scheme. An intense femtosecond pump pulse with a tuneable photon energy excites the sample, the magnetic response of which is monitored my measuring polarization rotation of a weaker probe pulse (photon energy 1.5 eV), delayed with respect to the pump. By changing the delay between the pump and probe pulses we are able to record the time evolution of the response. In this way we observe that in the borate $FeBO_3$ and the orthoferrites $TbFeO_3$, $ErFeO_3$ and $TmFeO_3$ the coherent spin oscillations are excited along with the coherent Raman phonons, when the photon energy of the pump pulse is tuned in resonance with an infrared-active optical phonon mode in these crystals. The phase and amplitude of the oscillations depends on the pump polarization, which is a signature of a coherent excitation mechanism. Our finding informs phono-magnonic engineering and provides invaluable insights into tuning pump photon energy (wavelength) to achieve maximal efficiency. Surpassing the previous demonstrations at cryogenic temperatures [2,3], we achieved coherent phonon-driven spin and lattice dynamics at room temperature and above, making the case for practical applications of the nonlinear phononics much stronger. In addition, in an insulating phase of the ferrimagnetic magnetite Fe_3O_4 we have discovered a new up-conversion nonlinear mechanism of exciting low-energy phonons by an intense THz pulse.

In summary, our results contribute to understanding the physics of ultrafast and coherent angular momentum transfer between light, lattice and ordered spins, which is a one of the key unresolved problems in magnetism.

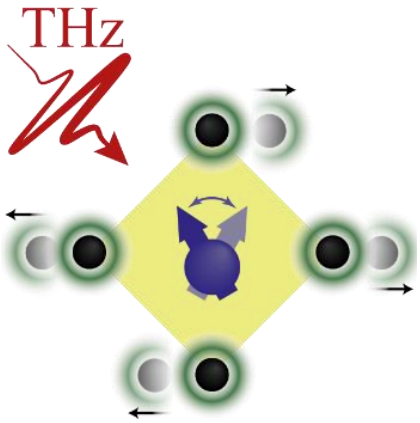

Fig. 1. The conceptual illustration of the phono-magnetism in iron oxides. A multi-THz ultrashort pulse (red) tuned in resonance with an optical phonon mode excites the magnetically ordered sample. Within a unit cell the phonon involves displacements (shown with black arrows) of the ions (black spheres) from their equilibrium positions such as the cell is distorted. This distortion leads to a sudden change of the microscopic magnetic couplings such as super-exchange and/or magnetic anisotropy that in turn exerts an impulsive torque onto the magnetic order (blue arrow), which starts to coherently oscillate at the frequency of a magnetic resonance.

[1] M. Först, et al. Nonlinear phononics as an ultrafast route to lattice control. Nature Physics **7**, 854 (2011).
[2] T. Nova, et al. An effective magnetic field from optically driven phonons. Nature Physics **13**, 132 (2017).
[3] D. Afanasiev, et al. Ultrafast control of magnetic interactions via light-driven phonons. Nature Materials **20**, 607 (2021).