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Ultrafast structural dynamics along a phase transition path

MnAs is a semi-metal that has attracted considerable interest since its discovery in the early 1900s. Current research on MnAs is motivated by its applications in spintronic devices or as a magnetocaloric material. The giant magnetocaloric effect of MnAs is directly related to an unusual sequence of magneto-structural phase transitions, the mechanisms governing these transitions not yet fully understood. Recent calculations suggest the involvement of a soft phonon (a mode of vibration whose frequency vanishes at the phase transition temperature). By using time-resolved diffraction measurements, an INSP team, in collaboration with LCLS researchers at Stanford, SOLEIL and ELETTRA synchrotrons, and the Beijing Semiconductor Institute, was able to identify this mode as an oscillation of atomic positions along the orthorhombic-hexagonal transition path of MnAs.

At low temperature, the most stable crystalline structure of MnAs is hexagonal and a ferromagnetic order (FM) is observed. This phase is commonly called α-MnAs. At T_c = 313 K, a first-order phase transition occurs, the FM order is lost and the structure becomes orthorhombic (β-MnAs). The orthorhombic distortion then decreases progressively and vanishes at Tt ≃ 400 K, where the hexagonal symmetry is recovered (γ-MnAs). Recent calculations suggest that the α-β transition is assisted by a soft mode, of frequency on the order of 1 THz, due to a strong spin-phonon coupling. This vibration mode would also be involved in the β-γ transition [1]. Until recently, there was no spectroscopic signature of MnAs phonons to test these predictions.

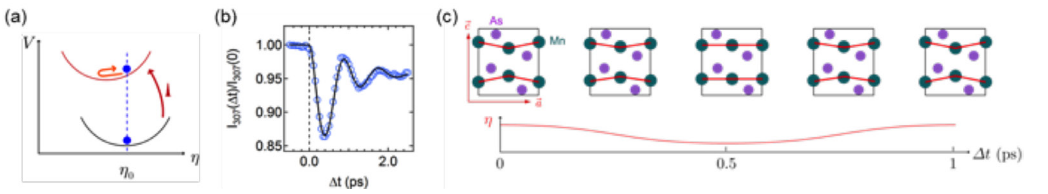

Figure 1
(a) Principle of the excitation of coherent phonons by a laser pulse, (b) Intensity of the 307 Bragg reflection as a function of pump-probe delay, (c) atomic movements corresponding to the probed mode within MnAs cell.

Coherent phonons, excited by laser in MnAs [Fig. (a)], could be detected using time-resolved X-ray diffraction [2]. Oscillations of the intensity [Fig. (b)] were measured for selected Bragg reflections with disctinct responses to the orthorhombic distortion. This confirmed that the probed phonon is indeed the predicted soft mode with atomic motions along the β-γ phase transition path [Fig. (C)]. The analysis of the measurements obtained by varying the distortion through the temperature revealed a softening of the mode. The experiments confirm the predictions, but only in part: the extrapolated frequency at zero distortion does not fall at zero. The β-γtransition is therefore not as simple as predicted (purely structural second-order transition).

These experimental results have thus made it possible to highlight the THz mode predicted by the theory and to confirm its strong softening when the orthorhombic distortion vanishes. They pave the way for new studies on spin-phonon coupling in the FM phase and on the role of this coupling in the β-γ phase transition.

References

[1] « Phonon Mechanism of the Magnetostructural Phase Transition in MnAs » J. Łażewski, P. Piekarz, J. Toboła, B. Wiendlocha, P. T. Jochym, M. Sternik, K. Parlinski, Physical Review Letters, 104, 147205 (2010)
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