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Phonons in thermoelectrics probed with neutron scattering experiments and DFT calculations: electron-phonon and phonon-phonon couplings in FeSi and PbTe.

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Thermoelectric materials are of broad interest for sustainable-energy applications, as they can convert waste heat into electricity, and provide solid-state refrigeration. Achieving high thermoelectric conversion efficiency requires limiting the thermal conductivity, through the disruption of phonon propagation. A detailed understanding of phonon dispersions and linewidths is thus critical in microscopic theories of thermal conductivity. We investigate the phonon dispersions and linewidths in thermoelectric materials for both refrigeration (FeSi) and waste heat recovery (PbTe), with an integrated approach combining neutron and x-ray scattering measurements with first-principles computer simulations. Our experiments benefit from the unprecedented neutron flux of the Spallation Neutron Source at Oak Ridge National Laboratory, to map phonon excitations throughout reciprocal space. The synergy between experiments and computer simulations is often a source of new insights into nature. We leverage powerful electronic structure calculations, including effects of finite temperatures, to analyze our experimental results. Our detailed investigations of the phonons and electronic structure in $Fe_{1-x}Co_xSi$ show that an adiabatic electron-phonon coupling leads to pronounced anomalies in the temperature dependence of both phonons and electron states [1]. The mechanism is general and could affect a broad class of materials. In PbTe, our measurements revealed a strong anharmonicity of the transverse-optic phonons, coupled to acoustic modes, providing a critical insight into the origin of the very low thermal conductivity in this rocksalt compound [2].

[1] O. Delaire, K. Marty, M. B. Stone, P. R. C. Kent, M. S. Lucas, D. L. Abernathy, D. Mandrus, and B. C. Sales, *Phonon softening and metallization of a narrow-gap semiconductor by thermal disorder*, Proc. Nat. Acad. Sci. USA **108**, 4725 (2011).

[2] O. Delaire, J. Ma, K. Marty, A. F. May, M. A. McGuire, M.-H. Du, D. J. Singh, A. Podlesnyak, G. Ehlers, M. D. Lumsden, and B. C. Sales, *Giant Anharmonic Phonon Scattering in PbTe*, Nature Materials **10**, 614 (2011).