OAK RIDGE NATIONAL LABORATORY
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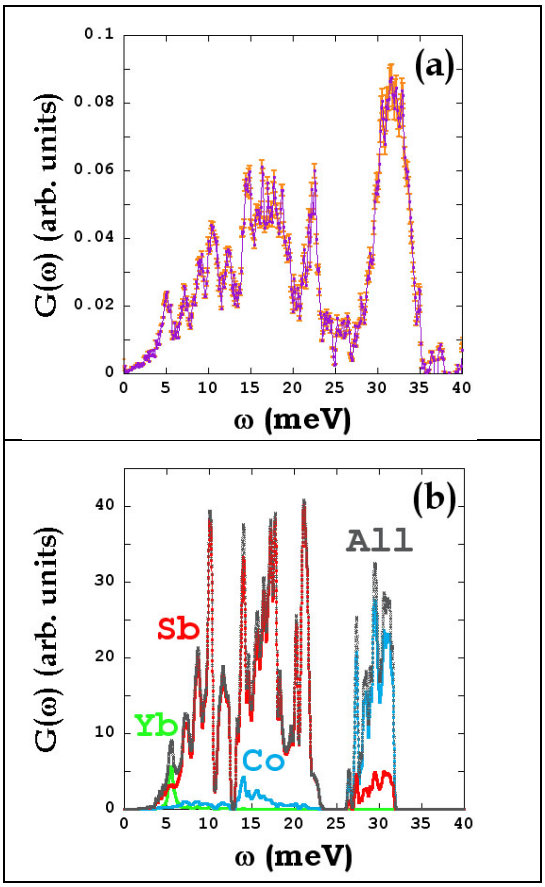
Neutron scattering reveals Einstein modes in the thermoelectric material $Yb_{0.2}Co_4Sb_{12}$

Researchers at ORNL's Spallation Neutron Source (SNS), in collaboration with the Condensed Matter Physics and Materials Science Department at Brookhaven National Laboratory (BNL), have investigated the properties of the thermoelectric material $Yb_{0.2}Co_4Sb_{12}$ and found evidence of Einstein oscillator modes at low and intermediate phonon energies.

Thermoelectric materials develop an electric potential in response to a temperature difference across their surfaces and are highly sought after for possible industrial use. This effect can be used in applications such as large-scale thermoelectric power generation from waste heat in power plants and cars, or for Peltier coolers (a type of refrigerator with no circulating fluid or moving parts). In such materials, both confinement effects and Einstein-like modes are desirable for improving the thermoelectric figure of merit, and both should be manifested as low-energy features in the measured phonon density of states.

Inelastic neutron scattering experiments carried out on the Cold Neutron Chopper Spectrometer at the SNS, combined with specific-heat measurements, provide compelling evidence for the existence of an Einstein oscillator (rattler) mode at ω_{E1}~5.0 meV in the filled skutterudite $Yb_{0.2}Co_4Sb_{12}$. Multiple dispersionless modes in the measured density of states of $Yb_{0.2}Co_4Sb_{12}$ at intermediate energy transfers ($14<\omega<20$ meV) are exhibited in both the experimental and theoretical density of states of the Yb-filled specimen. A peak at 12.4 meV is shown to coincide with a second Einstein mode at ω_{E2}~12.8 meV obtained from heat-capacity data. The local modes at these intermediate energy transfers are attributed to altered properties of the host $CoSb_3$ cage as a result of Yb filling. It is suggested that these additional dispersionless modes complement the low-energy Einstein mode at 5.0 meV in the scattering of heat-carrying phonons that is necessary to maintain a thermal gradient across the material. These observations offer a plausible explanation for the significantly higher thermoelectric figures of merit of filled skutterudites compared with their parent compounds.

Increasing the understanding of these intermediate energy modes is likely to demonstrate the importance of filler atoms in filled skudderite thermoelectric materials and allow the development of more efficient thermoelectric materials.



Shown is the remarkable correspondence between (a) the G(w) of $Yb_{0.2}Co_4Sb_{12}$ derived from the inelastic time-of-flight neutron-scattering experiment and (b) the results of the density functional perturbation calculation of the phonon DOS plotted in gray.

"Einstein modes in the phonon density of states of the single-filled skutterudite $Yb_{0.2}Co_4Sb_{12}$" I. K. Dimitrov, M. E. Manley, S. M. Shapiro, J. Yang, W. Zhang, L. D. Chen, Q. Jie, G. Ehlers, A. Podlesnyak, J. Camacho, and Qiang Li, *Physical Review B* **82**, 174301, November 2010.
http://prb.aps.org/pdf/PRB/v82/i17/e174301

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