Poster Presentation

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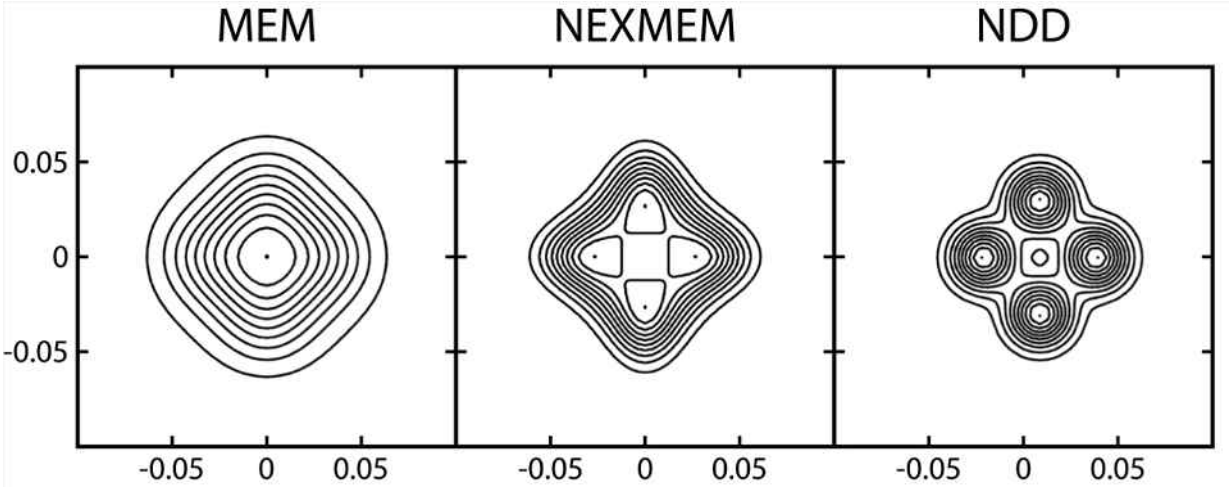
Characterization of Local Distortions in Thermoelectric Lead Chalcogenides

N. Bindzus[1], S. Christensen[1], M. Christensen[1], B. Iversen[1]

[1]*Aarhus University, Department of Chemistry and iNANO, Aarhus, Denmark*

Thermoelectric materials are functional materials with the unique ability to interconvert heat and electricity, holding much promise for green energy solutions such as efficient waste heat recovery. The extraordinary thermoelectric performance of binary lead chalcogenides has caused huge research activity, but the mechanisms governing their unexpected low thermal conductivity still remain a controversial topic. It has been proposed to result from giant anharmonic phonon scattering or from local fluctuating dipoles on the Pb site, emerging with temperature on the Pb site.[1,2] No macroscopic symmetry change are associated with these effects, rendering them invisible to conventional crystallographic techniques. For this reason lead chalcogenides were until recently believed to adopt the ideal, undistorted rock-salt structure. In the present study, we probe the peculiar structural features in PbX (X = S, Se, Te) using multi-temperature synchrotron powder X-ray diffraction data in combination with the maximum entropy method. Distorted atoms are detected and quantified by refinement of anharmonic probability density functions. The charge density analysis is complemented by nuclear density distributions (NDDs) reconstructed from neutron diffraction data and by a novel method: Nuclear Enhanced X-ray Maximum Entropy Method (NEXMEM). NEXMEM offers an alternative route to experimental NDDs, exploiting the superior quality of synchrotron X-ray data compared to neutron diffraction data. The increased atomic resolution introduced by NEXMEM proved essential for resolving atomic distortions, see figure below showing Pb in the (100) plane. Our findings outline the extent of disorder and anharmonicity in binary lead chalcogenides, promoting our fundamental understanding of this class of high-performance thermoelectric materials. The applied approach can be used in general, opening up for widespread characterization of subtle features in crystals with unusual properties.

[1] *E. S. Bozin et al., Science, 2010, 330, 1660-1663,* ***[2]*** *O. Delaire et al., Nature Materials, 2011, 10, 614-619*



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