Atomic dynamics in superionic argyrodite Ag_8SnSe_6

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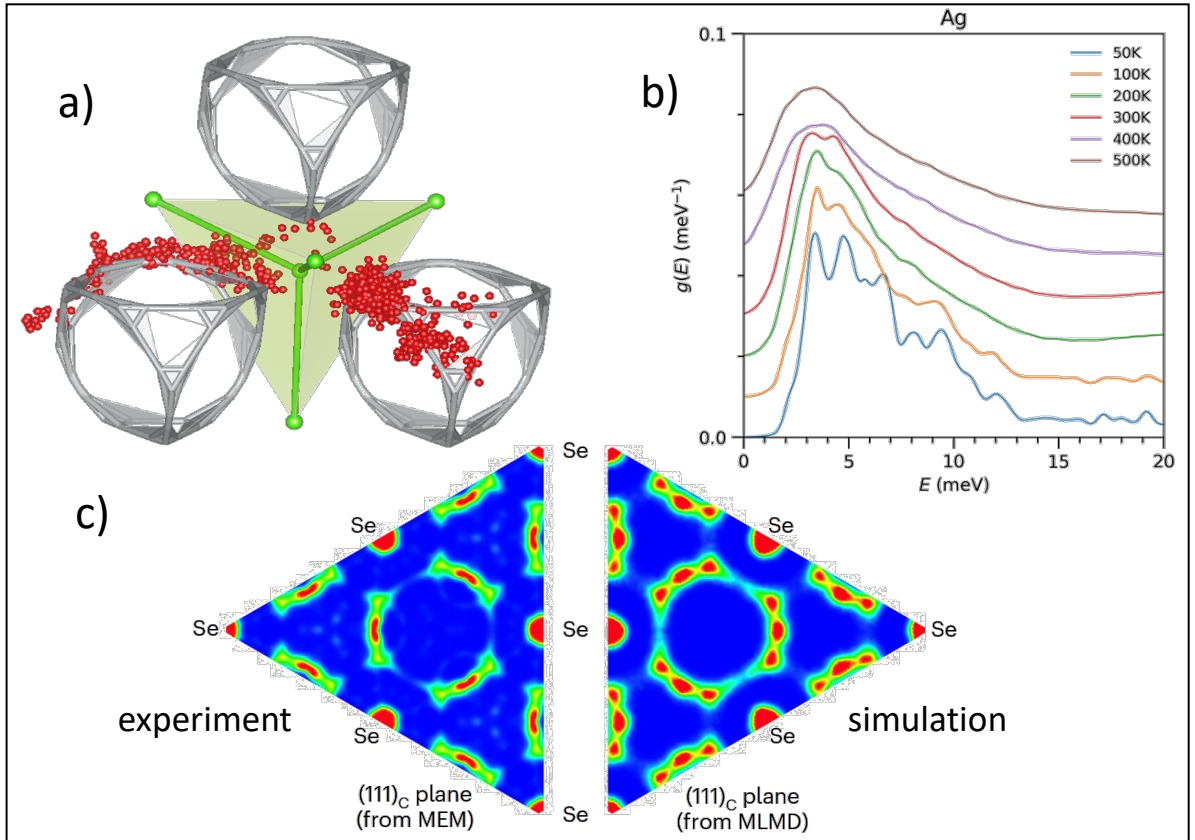
Superionic materials are solids in which mobile cations diffuse as fast as in liquids, enabling excellent performance both as solid-state electrolytes for batteries and as thermoelectric converters. In the spirit of MGI, we integrated atomistic simulations with experiments to shed light into the complex atomic dynamics of superionics.

We investigated ionic diffusion and lattice vibrations in the argyrodite compound Ag_8SnSe_6 using first-principles simulations extended with machine-learning based molecular dynamics and compared results with x-ray and neutron scattering measurements. We identified a critical interplay of mobile Ag^+ motions and host framework vibrations that enables the facile hopping of Ag^+ ions.

The detailed atomistic simulations showed the importance of extreme anharmonicity and corrugation of the potential energy landscape, which is reflected in the strong temperature dependence of phonon vibrational spectra. This underpins both the ultralow thermal conductivity and fast ionic diffusion in this compound. Our results provide fundamental insights into the complex atomic dynamics in superionic materials for energy conversion and storage.

Q Ren, MK Gupta, M Jin, J Ding, ... Y Pei*, O Delaire*, J Ma*, "Extreme phonon anharmonicity underpins superionic diffusion and ultralow thermal conductivity in argyrodite Ag8SnSe6", **Nature Materials 22**, 999 (2023).
DOI: https://doi.org/10.1038/s41563-023-01560-x



Simulations of atomic dynamics in the superionic argyrodite Ag_8SnSe_6. (a) Computed trajectory showing Ag^+ (red) hopping between silver cluster through a bottleneck area modulated by positions of green Se atoms. (b) Strong broadening of Ag vibrational spectrum upon warming. (c) measured (left) and computed (right) probability densities in superionic phase.