



Contribution ID: 60

Type: **Contributed Oral**

Li-ion Dynamics in Solid Electrolytes: An Integrated Neutron-based techniques and Terahertz Time-Domain Spectroscopy Study

Tuesday, 10 February 2026 15:15 (15 minutes)

All-solid-state batteries (ASSBs) hold significant commercial promise, driven by their inherent potential to deliver exceptional energy density, power density, and safety. While factors such as rate capability, interfacial stability, and extended cycle life hinder the development of ASSBs, which are critically dependent upon ionic conductivity enhancements within solid-state electrolytes (SSEs). Among SSE families, $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) demonstrates exceptional commercialization potential due to its intrinsic compatibility with lithium metal and outstanding thermal stability under ambient conditions. However, in Li-ion SSEs, the intricate Li⁺ migration dynamics remain inadequately understood, critically constraining rational materials design.

In this study, Nb mono-doped and Ta/Nb equimolar co-doped LLZO samples ($\text{Li}_{6.4}\text{La}_3\text{Nb}_{0.6}\text{Zr}_{1.4}\text{O}_{12}$ and $\text{Li}_{6.4}\text{La}_3\text{Ta}_{0.3}\text{Nb}_{0.3}\text{Zr}_{1.4}\text{O}_{12}$, respectively) were fabricated via spark plasma sintering (SPS). Implementing neutron powder diffraction (NPD) at ANSTO, we have obtained Li-ion-centric crystallographic information, which is more reliable than conventional lab-based X-ray powder diffraction. Inelastic neutron scattering (INS) revealed the energy distribution, population, and Q-dependence of Li-centered vibrational modes, as well as the momentum-integrated Li-dominated phonon density of states (DOS) that controls the attempt frequency and many-body phonon-ion coupling, which can only be isolated from the heavy La-O/Zr-O framework with neutron-based probes.

To reveal complementary microscopic mechanisms and to probe Li⁺ ions' transient pre-hopping behaviors, we implemented in situ THz time-domain spectroscopy (THz-TDS) to drive ultrafast on-site Li⁺ ions vibrating at their intrinsic oscillation frequencies ($\sim 10^{12}$ - 10^{13} s⁻¹). Fourier transforming spatiotemporal resolved spectra, we yielded frequency spectra, then computed the THz conductivity that is dominated by localized vibrational modes coupled to polar lattice phonons.

Overall, this study serves as a proof of concept for utilizing Neutron-based and THz-TDS to probe ion dynamics in ionic conductors. Also, we have shown the scientific workflow to delve the mechanistic insights with these techniques.

Field of Condensed Matter

Energy and Functional Materials

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Session Classification: Spectroscopies 1

Track Classification: Contributed talk sessions: Spectroscopies