Experimental visualization of conduction pathways and direct observation of fast lithium-ion diffusion in superionic conductors using neutron scattering

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Designing the next generation of lithium-ion batteries (LIBs) requires fundamental information that surprisingly often is still unknown – for example, details of how ions actually move through a working device. Li$_2$S–P$_2$S$_5$ superionic conductors have received a great deal of attention as a solid electrolyte for all-solid-state LIBs because of its extremely high ionic conductivity of 10$^{-3}$ to 10$^{-2}$ S/cm at room temperature [1,2]. The authors predict and visualize the conduction pathways of lithium ions in Li$_2$S–P$_2$S$_5$ glasses and Li$_7$P$_3$S$_{11}$ crystal, using reverse Monte Carlo (RMC) modelling [3] and the bond valence sum (BVS) approach [4] with neutron and synchrotron X-ray diffraction data (see Fig. 1(a)). The conduction pathways of lithium ions can be classified into two types: relatively “stable” regions and “metastable” regions for lithium-ion diffusion, respectively [5]. Moreover, the authors use quasi-elastic neutron scattering (QENS) to directly monitor the fast diffusion of lithium ions in Li$_7$P$_3$S$_{11}$ (see Fig. 1(b)). According to the jump diffusion model, the lithium ions migrate between stable regions within a jump length $<l> = 4.3$ Å along the conduction pathways [6]. This understanding is key to improved energy storage for applications ranging from vehicles to further smart grids.

Figure 1. (a) Conduction pathways of Li ions in Li$_7$P$_3$S$_{11}$ superionic conductor at room temperature, predicted by RMC modelling and the BVS approach. (b) QENS spectra of Li$_7$P$_3$S$_{11}$ ($Q = 0.32$ Å$^{-1}$) collected at 150, 297, and 473 K.