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ABSTRACT

Ultrafast Li-Ion Dynamics in the Solid Electrolyte $LiTi_2(PS_4)_3$ as Resolved by NMR Reaching Cryogenic Temperatures

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Self-diffusion processes of small atoms or ions play a crucial role in many areas of research. The unique crystal structure of $LiTi_2(PS_4)_3$ (LTPS) presents a variety of energetically in-equivalent diffusion pathways for small Li^+ charge carriers and has resulted in one of the highest Li^+ diffusion coefficients. Investigating these pathways individually at the atomic scale poses significant challenges, especially for probing jump processes. In this study, we utilized nuclear spin relaxation techniques down to cryogenic temperatures to reveal unprecedented details about both long-range and short-range Li^+ dynamics. The temperature-dependent ^7Li NMR spin-lattice relaxation rate exhibits a series of diffusion-induced peaks, allowing the extraction of activation energies and jump rates. Due to the exceptionally fast localized Li^+ exchange processes in LTPS, temperatures as low as 20 K are required to freeze Li^+ dynamics entirely within the ring-like cages of the LTPS structure. Several NMR signatures might suggest that, in addition to classical over-the-barrier hopping, quantum tunneling of ^7Li spins may contribute to the ultrafast dynamics observed.