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Resolving Grain Boundary Microstructures in Garnet-Type $Li_7La_3Zr_2O_{12}$ using Model-Based TEM Image Simulation

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Solid-state batteries have been at the forefront of energy research due to the advantages in safety and high packing density for large-scale and long-term applications. As a key component of solid-state batteries, solid electrolytes have attracted a lot of attention in the past years. Intensive research in this field is directed at improving the ionic conductivity of ceramic solid electrolytes through materials advancement and microstructure engineering. Li7La3Zr2O12 (LLZO) is a garnet-type oxide ceramic electrolyte material that possesses promising properties with a good balance between ionic conductivity and structure stability [1]. It is noted that defects, including grain boundaries, micropores, and other local imperfections, play a vital role in affecting the ionic conductivity through the modulation of the fundamental structure-property relationship. Therefore, it is essential to characterize the defects and reveal their impact on ionic conductivity in the LLZO system.

Transmission electron microscopy (TEM) is a powerful tool for the characterization of materials structures and defects on the atomic scale [2]. However, TEM only provides two-dimensional projection images, which is insufficient to conclusively resolve the entire characteristics at all degrees of freedom. Here, we intend to leverage the prior crystallographic knowledge of the target LLZO system and use the model-based image simulation approach to resolve grain boundary microstructures with representative features from individual atomic-resolution images. Figure 1a shows the unit cell of garnet-type LLZO in the cubic lattices, consisting of a complex skeleton with oxygen-coordinated tetrahedra, octahedra, and dodecahedra. In a case study shown in Figure 1b, the atomically resolved high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image contains a sharp grain boundary. By analyzing the lattice orientations on both sides of the grain boundary, we have obtained the necessary information to build an atomistic model through a trial-and-error manner, as shown in Figure 1c. In this model, two grains are aligned along [001] and [111] axes with a shared {110} plane to coherently separate in between. Using the multislice STEM image simulations via Dr. Probe software [3], we reproduced the HAADF-STEM image consistent with the experimental data, which confirms that the proposed grain boundary model is validated. This work offers a rational approach to resolve defect structures in complex crystal lattices, which is a necessary step to understand the fundamental structure-property relationship in functional material systems. In conjunction with the Microscopy Society of America Undergraduate Research Scholarship, we also demonstrate that the model-based simulation is an effective toolset for advancing educational effectiveness in the field of materials science and engineering [4].

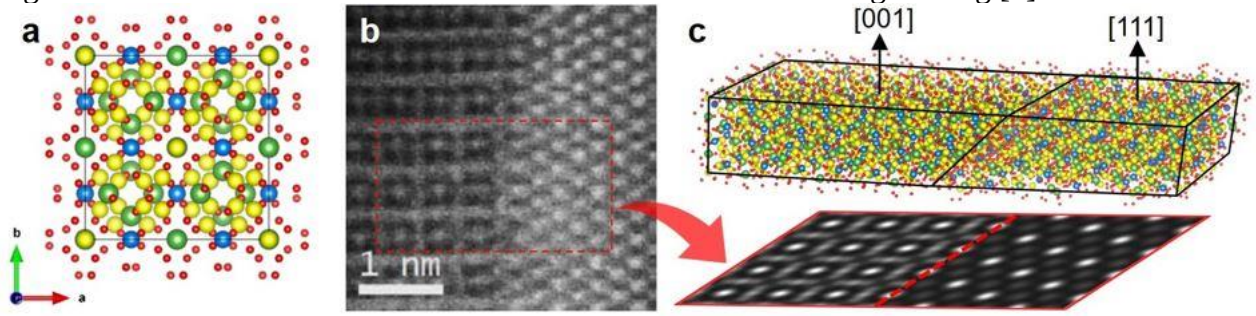

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Figure 1. (a) Unit cell of cubic-LLZO lattice. (b) HAADF STEM image of LLZO containing a grain boundary. (c) Multislice STEM image simulation based on the atomistic model of a two-grain microstructure.

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