Bridging Theory and Experiment: Point Defect Characterisation in Doped $SrTiO_3$

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Defect engineering is a pivotal strategy for tailoring the electronic and magnetic properties of complex oxides. However, bridging the gap between microscopic point defect properties and macroscopic material behavior remains a significant challenge. To establish this link, first-principles modelling via density functional theory (DFT) is essential, as it provides the necessary structural and electronic foundation to interpret advanced microscopy measurements and predict bulk functionality. While crucial for optimizing performance, the reliable characterization of individual defects in the bulk is often hindered by limitations in spatial resolution, signal sensitivity, and experimental stability, necessitating the development of new experimental strategies. Addressing these experimental requirements, we performed atomic-resolution 4D-STEM experiments on $SrTiO_3$ doped with 0.5 wt% tantalum (Ta) using a Nion HERMES microscope operating at 60 kV with a semi-convergence angle of 36 mrad. Meticulous optimization of experimental conditions and the preparation of exceptionally thin specimens, limited to only a few nanometres in thickness, were essential to ensure the detection of subtle defect signals.[1] We employ DFT calculations[2] to determine the relaxed atomic structures of relevant point defects, such as Ta substitutional dopants and associated Sr vacancies. These DFT-derived geometries serve as input for multislice simulations of electron scattering[3], enabling the prediction of defect-induced modifications to the angular scattering distribution.[4] We demonstrate that these sub-angstrom atomic displacements produce characteristic diffuse scattering contributions (Huang scattering[5]), which significantly influence the momentum-resolved signal measured in 4D-STEM.[6] By incorporating these structural distortions into the simulations, we identify defect-specific scattering signatures and design optimized virtual detectors to enhance sensitivity to individual point defects. By combining DFT-derived defect structures, multislice simulations, and 4D-STEM, we establish a robust framework for the detection and characterization of individual point defects. This integration of first-principles modelling with advanced electron microscopy provides a pathway toward quantitative defect analysis in complex oxides and other functional materials.

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