

Materials Science and Engineering Dissertation Defense

Probing Local Characters of Nanoparticles with Monochromated Electron Energy-Loss Spectroscopy

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Abstract

Electron energy-loss spectroscopy (EELS) combined with scanning transmission electron microscopy (STEM) enables materials characterization with high spatial resolution. Advances in electron optics now allow the investigation of low-energy excitations with high energy resolution, including optical and vibrational properties, which are essential for understanding energy conversion processes. The objective of this work is to refine and expand EELS methodologies and apply them to the study of heterogeneous catalytic nanoparticles.

Aloof-beam EELS is an effective technique for probing low-energy material properties, e.g., optical and vibrational properties, at a distance, thereby avoiding radiation damage. However, the spatial distribution of energy deposition in the material under this configuration, which are the source of the measured signal, is not fully understood. Finite-element dielectric simulations were employed to explore this energy deposition profile as a function of experimental parameters, including dielectric functions, impact parameters, beam energy, and energy loss.

To achieve a deeper understanding of the photonic response of nanostructures, such as photonic modes in CeO₂ dielectric nanocubes, monochromated EELS was used. EELS mapping revealed that photonic modes exhibit distinct spatial excitation preferences and are sensitive to variations in the surrounding environment. Dielectric simulations corroborated these observations and visualized the modes by mapping the internal electric fields.

A significant limitation of monochromated EELS is its low signal-to-noise ratio (SNR). To address this challenge, the unsupervised deep video denoiser (UDVD), originally developed for denoising low-dose TEM images, was adapted to EELS spectral mapping datasets. The neural network was modified for spectroscopic data and demonstrated to effectively enhance SNR in both simulated and experimental datasets.

In thermocatalytic reactions, phonon and vibrational behavior near catalyst surfaces are hypothesized to play a critical role in determining reaction behaviors. Off-axis vibrational EELS was employed to probe vibrational behavior in catalytic nanoparticles, including NiO and CeO₂, with atomic resolution. Distinct changes in vibrational modes were observed in the surface regions, indicating unique atomic vibrational dynamics near nanoparticle surfaces.

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