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Simulation of STEM-EELS Including Diffraction and Solid-State Effects II: Adding the Experiment.

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Aberration corrected scanning transmission electron microscopy (STEM) has provided the ability to measure electron energy loss near edge structure (ELNES) at atomic resolution. Most current simulations of ELNES assume a dipole approximation and ignore the diffraction of the incident electron by the sample. Here we combine a non-dipole density-functional calculation of the mixed dynamical form factor (MDFF) [1] with diffraction theory to describe the generation of NES in the aberration corrected STEM. As a model system for the initial application of the theory, we use $LaMnO_3$ which has been shown to have two nonequivalent oxygen sites, which we label O1 and O2 [2].

The inelastic scattering coefficient describing ELNES, for an incident electron of wave vector $\mathbf{k}$, may be written as

$$\frac{\partial \mu_{\mathbf{h},\mathbf{g}}}{\partial E} \equiv \mu'_{\mathbf{h},\mathbf{g}} = \frac{1}{2\pi k V_c} \sum_n \exp\left[-M\left(\mathbf{g}-\mathbf{h}\right)\right] \exp\left[2\pi i \left(\mathbf{g}-\mathbf{h}\right)\cdot \boldsymbol{\tau}_n\right] F\left(\mathbf{h},\mathbf{g}\right)$$

where $\boldsymbol{\tau}_n$ describes the atomic locations within the unit cell of volume V_c, and M is a Debye-Waller factor. The quantity $F\left(\mathbf{h},\mathbf{g}\right)$ is written in terms of the MDFF as

$$F\left(\mathbf{h},\mathbf{g}\right) = \frac{1}{2\pi^3 a_0^2} \int_{\text{detector}} k' \frac{S\left(\mathbf{q}+\mathbf{h},\mathbf{q}+\mathbf{g}\right)}{\left|\mathbf{q}+\mathbf{h}\right|^2 \left|\mathbf{q}+\mathbf{g}\right|^2} d\Omega.$$

Here $\mathbf{k}'$ is the wave vector of the scattered electron, $h\mathbf{q} = h\left(\mathbf{k}-\mathbf{k}'\right)$ is the momentum transfer to the crystal.

In Fig. 1 we examine the role of the detector size in determining the shape of the NES for $\mu'_{\mathbf{0},\mathbf{0}}$. We have assumed incident electron energy of 60 keV. Intensities have been normalized to a maximum of one for ease of comparison. For the O2 site, there is very little variation in the NES as a function of detector size. However the NES of the O1 site changes significantly as the detector increases in size. The ratio of the first two peaks varies rapidly as the detector size is increased and, for large detectors, these peaks dominate the higher energy loss structures. This can be explained in terms of the different localizations of the inelastic scattering transitions to different final states. We examine the role of detector geometry by considering the wave function of the inelastically scattered electron.

The $\mu'_{\mathbf{0},\mathbf{0}}$ term does not however tell us the whole story; it assumes plane wave illumination and the absence of diffraction. In Fig. 2 we examine the form of the inelastic scattering coefficients for the first few elements in the scattering matrix. The diagonal terms are almost identical for the 30 mrad detector semi-angle used here. The off-diagonal elements show significant variation, and since these elements are displayed on an absolute scale, their contribution is significant, especially for atomic resolution STEM, where the approximation of plane wave illumination is poor.

In addition, diffraction of the incident probe by the sample leads to higher order coefficients (beyond the range of the probe forming aperture) being excited. It should be noted that the diagonal terms do not contain site information, and the off diagonal terms in the MDFF are an essential part of the physics describing the generation of ELNES [3].

References

[1] M.P. Prange et al., *Microsc. Microanal.* (2011) proceedings of this conference.
[2] M. Varela et al., *Phys. Rev.***B**. 79 (2009) 085117.
[3] This research was supported by DOE grant DE- F002-09ER46554 (MPP, MPO, STP), and the Office of Basic Energy Sciences, Materials Sciences and Engineering Division, U.S. Department of Energy (SJP).

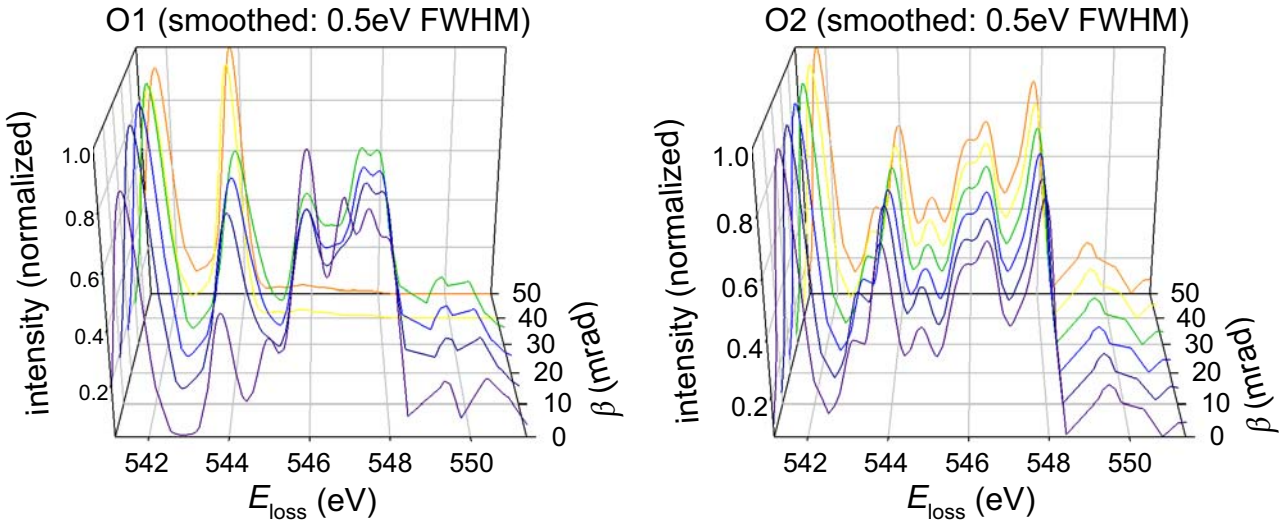

FIG. 1. Normalized intensity of the dynamic form factor integrated over detector semi-angle β for each of the nonequivalent oxygen sites in $LaMnO_3$. Simulations are for an incident energy of 60 keV.

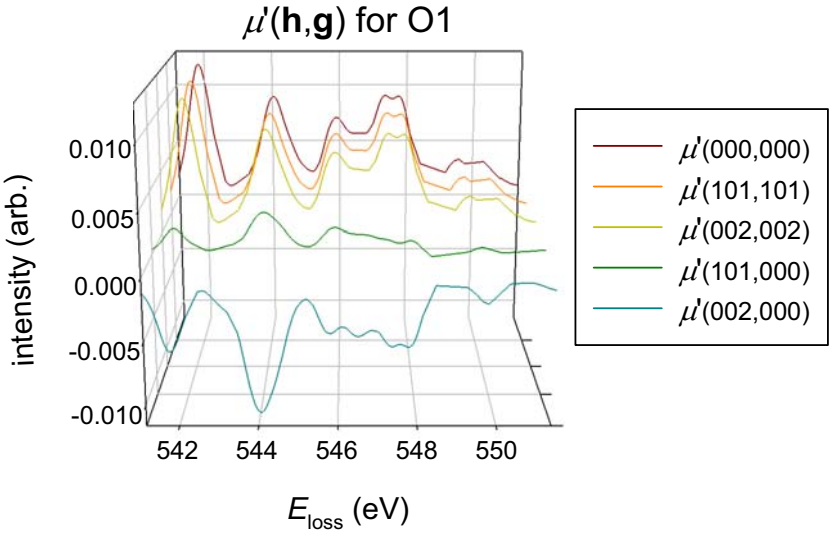

FIG. 2. Contributions from the diagonal and off-diagonal inelastic scattering coefficients for O1 in $LaMnO_3$ oriented in the 010 zone axis orientation. Simulations assume an incident energy of 60 keV and a 30 mrad collection semi-angle for the EELS spectrometer. The real parts of the complex coefficients are shown.

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