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Real-space Measurements of Bonding Charge Density in Aberration-corrected HREM

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Measurement of valence charge density, the distribution of electrons important to bonding, is a rather well established technique for bulk materials within the diffraction community using a variety of techniques: single crystal x-ray diffraction, powder x-ray diffraction, convergent-beam electron diffraction and recently precession electron diffraction. All of these reciprocal-space techniques have the advantage of high positional accuracy, but the partitioning of bonding electrons when fitting diffraction data is a non-convex problem that can lead to divergently stable solutions with regard to local minima if one is not extremely careful.

The improvements in resolution and image interpretability due to the introduction of spherical aberration correctors have now enabled the pursuit of structural bonding information in direct-space. Recent high-resolution transmission electron microscopy (HREM) multislice simulations utilizing the fully relaxed potential form density functional theory (DFT) calculations have shown that for light element oxides, valence electron effects can contribute as much as 24% of the total contrast in HREM images [1]. We have taken this analysis further by more fully exploring the thickness and defocus parameter space trough image simulation utilizing parameters for an aberration-corrected JEOL-2200MCO microscope. Additionally, exit wave restorations of simulated defocus series were performed and the resulting amplitude and phase images were quantified for sensitivity to bonding effects (Fig1). It was found that the amplitudes of the restored exit waves were more sensitive to subtle bonding effects than were the phases and that the optimal thickness for observation of bonding was ~20-25 nm (Fig 2).

To investigate these findings experimentally, we have performed measurements of bonding effects in energy-filtered images along the [010] projection of the mineral Forsterite (Mg_2SiO_4) using the aberration-corrected JEOL-2200MCO microscope at the University of Oxford. Focal series exit wave restorations were performed on the experimental image data and compared with similar restorations of simulated images computed from both traditional isolated-atom scattering factors and fully-bonded structures. The experimental findings will also be compared with full-potential all-electron DFT calculations of the projected charge density of Forsterite.

[1] B. Deng, and L. D. Marks, Acta Crystallogr. Sect. A **62**, 208 (2006).

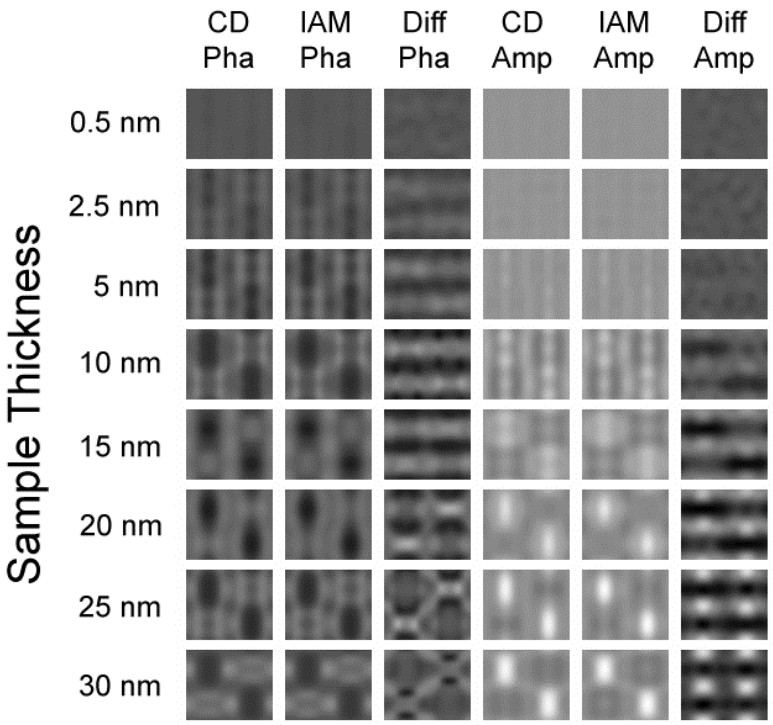

Figure 1: Amplitude and phase of exit waves restored from simulated focal series under C5-limited imaging conditions with isolated atom (IAM) and full DFT charge density (CD) potentials. Contrast in difference images increased 10x.

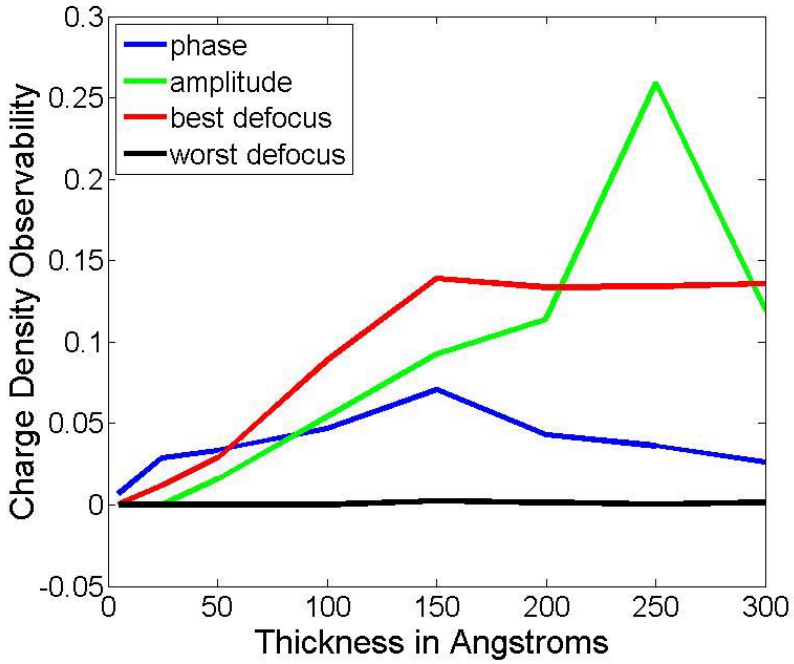

Figure 2: Observability metric of the amplitude and phase of exit waves restored from simulated focal series. Also included in the plot are the best and worst values for any single image in the focal series at each thickness.

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