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Spectral imaging for characterization of complex defect structures using *ab initio* ELNES

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Aberration corrected scanning transmission electron microscopy (STEM) has become a modern experimental technique for the characterization of materials with defects, interfaces, and microstructures. High resolutions of up to 0.5 Å can be achieved that clearly resolve atomic images in the presence of defect structures such as dopant atoms in buried interface [1], segregated ions to grain boundaries [2], interface structures of intergranular glassy films (IGF) [3], etc. We have recently developed a theoretical spectral imaging (SI) technique [4] that can be a powerful complement to the experimental probe. In this method, the defect structure under investigation is modeled by a large supercell of up to several hundred atoms. The atom and edge specific electron energy-loss near edge structure (ELNES) (or XANES) spectra of *every* atom in the model are calculated using the *ab initio* OLCAO method [5]. This is carried out by "exciting" a core electron in the target atom with the core-hole interaction included. The atom-centered spectra from all the atoms in the model are then collected and interpolated onto a dense 3D mesh. A 4-dimensional functional field $P(\mathbf{r}_i,E_n)$ at each point $\mathbf{r}_i$ and energy scale n is defined. A Euclidean difference $D_i(P_0,P_i)$ is then evaluated for every mesh point where the difference is between the functional field spectrum at the mesh point $\mathbf{r}_i$ and an arbitrary reference spectrum $P_0(\mathbf{r}_i,E_n)$ (say the O-K edge of an O atom in a defect-free part of an O containing model). $D_i(P_0,P_i)$ is a scalar field which can be visualized in any direction at any angle. Images of the variations in the atomic interaction can be obtained by a judicious choice of the energy range E_n in ELNES because different ranges reflect different orbital interactions,

Figs. 1 and 2 show the theoretical SI of a model containing a passive defect in Si with 5-, 6-, 7-, and 8-membered rings but no broken bonds. Such passive defects are very difficult to detect experimentally. The theoretical SI is able to show changes in the spectra due to the presence and structure of the passive defect for various different energy ranges of the Si-K edge. New SI results on the O-K edges of a 240-atom model of a Σ5 grain boundary in $SrTiO_3$ and the N-K edges of N atoms in the glassy region of a 907-atom prismatic IGF model in β-Si_3N_4 will also be presented. Possible applications and further improvements on this theoretical SI technique will be discussed.

References

[1] N. Shibata, et al., Nature Materials, 8, 654-658 (2009).
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[3] N. Shibata, et al, Nature 428, 731-733 (2004).
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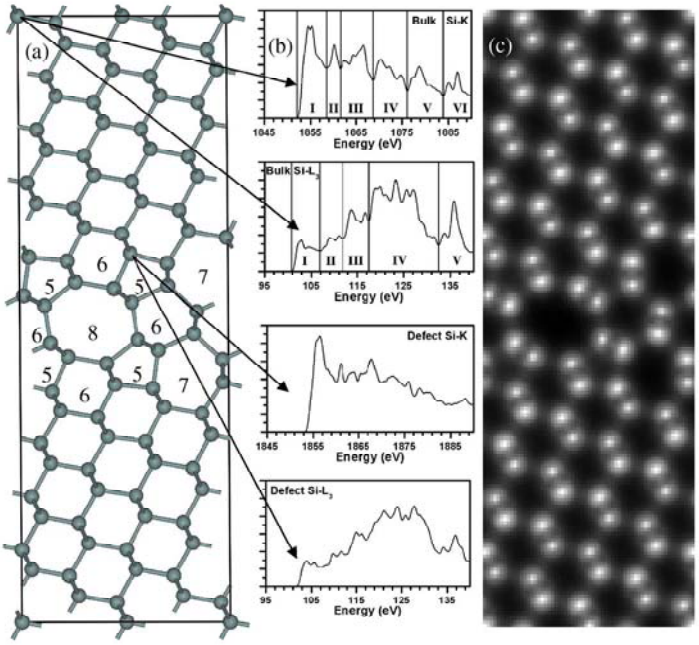

Fig. 1(a) Model of the 180-atom planar defect model in Si (a=12.592 Å, b=7.593Å, c=36.625 Å, β=89.656º) showing different ring structures. (b) Calculated Si-K and Si-L_3 edges at two locations: one in the bulk region and one in the defect region. Si-K and Si-L_3 edge spectra are divided into energy windows which are the identified features for the function field for different SI. (c) Simulated ADF STEM image for this model with the following parameters: 6 layers (about 4.5 nm) thick, convergence angle 22 mrad, all aberrations set to zero, and inner detector angle set to 25 mrad at 300 kV.

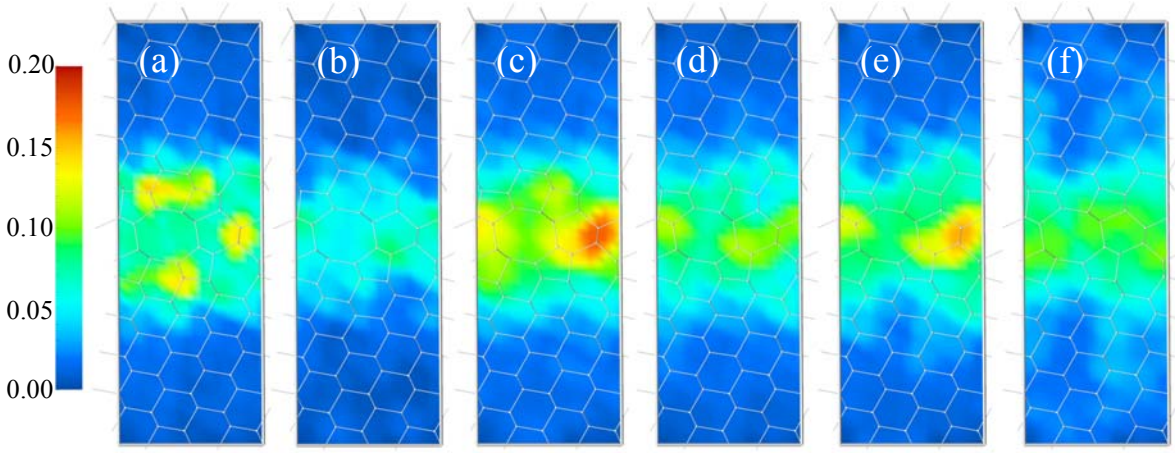

Fig 2. SI (a) to (f) use regions I to VI of the Si-K edge in Fig. 1(b) as the function field sources with shading (colors) higher on the scale indicating a greater degree of deviation from the bulk region spectrum.