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A menu of electron probes for optimising information from scanning transmission electron microscopy

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ABSTRACT

We assess a selection of electron probes in terms of the spatial resolution with which information can be derived about the structure of a specimen, as opposed to the nominal image resolution. Using Ge [001] as a study case, we investigate the scattering dynamics of these probes and determine their relative merits in terms of two qualitative criteria: interaction volume and interpretability. This analysis provides a 'menu of probes' from which an optimum probe for tackling a given materials science question can be selected. Hollow cone, vortex and spherical wave fronts are considered, from unit cell to Ångstrom size, and for different defocus and specimen orientations.

1. Introduction

With recent improvements in electron optics, atomic resolution scanning transmission electron microscope (STEM) images and spectroscopic maps have become readily achievable. However, possessing atomically-resolved features does not guarantee an image provides atomic resolution information [1–7]. Electron probes focused to an atomically-fine cross-over scatter such that the signal from a given probe position may include contributions from a sample volume extending well beyond the column beneath the probe [1–7]. Fig. 1 illustrates this point. In STEM images that resolve atomic columns, it can be tempting to assume the signal arises from only those atoms in the column upon which the probe is placed (Fig. 1(a)). However, in all but the thinnest of samples the probe scatters beyond its initial impact point, ensuring an appreciable contribution from atoms in adjacent columns (Fig. 1(b)). This raises two interrelated questions: How can we tune the electron probe to optimise the *information* resolution, irrespective of the nominal *image* resolution? And how can we best tailor the probe to control the spatial origin of the ADF-STEM signal and optimise interpretability?

In this work, we present a 'menu' of electron probes and assess their relative merits. Each probe delivers a different scattering dynamic and spatial sampling, from which the most suitable probe for a given task can be selected. In particular, we assess this suitability using two qualitative concepts – 'interaction volume' and 'interpretability', to be defined presently – with the nominal spatial resolution of the image being of less importance.

By 'interaction volume' we mean the volume of specimen that has scattered the electron probe with sufficient 'weight' to make a significant contribution to the ADF-STEM image. In Fig. 1, those atoms deemed to make a significant contribution are shaded (darker shades mean greater contribution), and the dashed lines indicate the boundary of the interaction volume containing these atoms. In the scenario in Fig. 1(a), the interaction volume is a single atom wide, matching the nominal resolution of the incident probe and ADF-STEM image. In the scenario in Fig. 1(b), the spreading of the electron probe through the crystal leads to atoms from several atomic columns being sampled. As such, the interaction volume is nanoscale and the likely information resolution is much lower than the image resolution.

By 'interpretability' we mean the degree to which the shape of the interaction volume, and the distribution of scattering weight within it, can be estimated without detailed scattering calculations. The scenario depicted in Fig. 1(a) supposes that each atom in the column contributes to the signal with equal weight, regardless of its position in the column. If achievable, this would enable a straight forward interpretation providing information with atomic resolution. It would be particularly helpful for certain materials problems, such as identifying the type and (lateral) position of single atomic defects in crystals. In the scenario depicted in Fig. 1(b), each atom within the volume contributes to the ADF-STEM image

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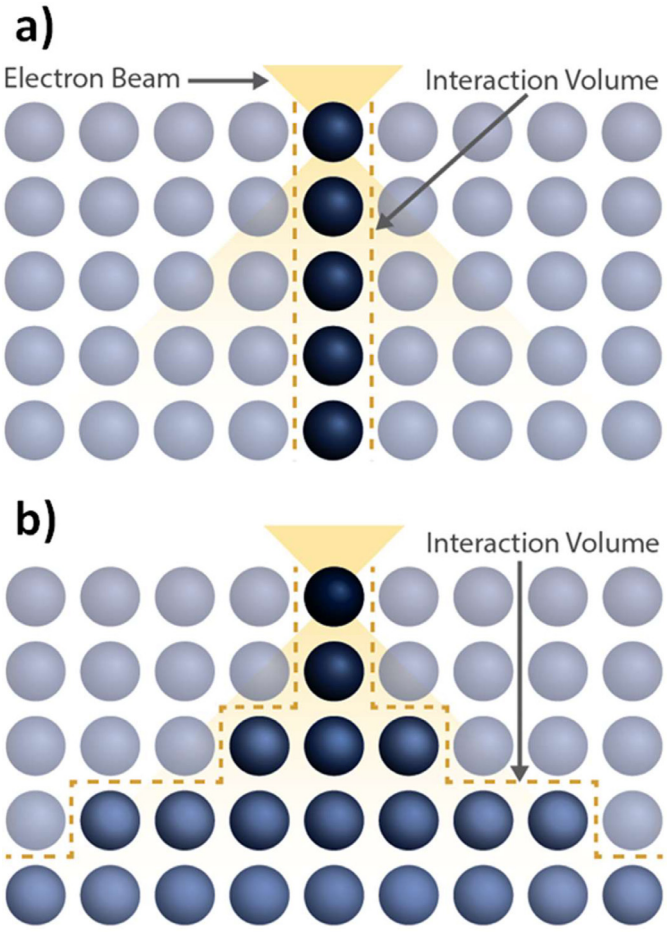

Fig. 1. The interaction volume is defined by the volume of the specimen that has scattered the electron probe with sufficient 'weight' to make a significant contribution to the ADF-STEM image. The boundary of the interaction volume is indicated with the dashed line. The relative weight with which scattering from different atoms contributes to the ADF-STEM image is indicated here by the tonal difference of the spheres (deeper shade corresponding to stronger signal contribution). Two different scenarios are shown: (a) channelling along a single column beneath the probe position and (b) scattering beyond the column beneath the probe.

with a different weight, adding to the complexity of interpretation (unless the structure is homogeneous and/or the interaction volume and scattering weight could somehow be known *a priori*).

There have been many studies [2–11] investigating the effect of various parameters on the scattering dynamics of the probe which have informed our choice of which parameters to investigate here, namely convergence angle, probe defocus and specimen orientation. While larger convergence angles give better STEM image resolution compared to smaller angles [12], they also result in a larger lateral dispersion of the probe [5], exacerbating the potential disconnect between image resolution and information resolution. Defocusing the probe and tilting the specimen away from major zone axes have been identified as a means to mitigate 'channelling' effects of the probe [13–16] – in this context, channelling has been used to mean the tendency for the probe electrons to travel preferentially down atomic columns [17] – and are thus anticipated to aid interpretability. We also investigate hollow cone [18] and vortex [19] probes, which offer intrinsically different scattering dynamics [10,20].

Unfortunately, the complexity of scattering is such that no single probe configuration considered here is unilaterally better than the others in terms of interaction volume and interpretability. However, we emphasize the relative strengths and weaknesses of the different configurations for certain materials applications and scenarios.

2. Calculating the contribution of each atom to the ADF-STEM signal

To understand the spatial resolution of the information contained within an ADF-STEM image, we calculate the spatial origin of the signal reaching the ADF detector. It is important to note that the spatial origin of the signal is a consequence of - but not the same as - the extent of the scattered electron wave function within the crystal (the latter having been studied previously under a variety of conditions [2–6,10]).

All calculations assumed an aberration-free 300 kV probe. The spatial coherence function of the probe was described using a Gaussian with a FWHM of 0.8 Å. An ADF detector with an inner angle of 50 mrad and an outer angle of 200 mrad was used. Ge [001] was used as the test case since all atomic columns are identical, which serves to isolate the effect of ADF-STEM signal mixing from other effects which may arise from a difference in atomic number. Multislice calculations were carried out on a supercell comprising a 6×6 tiling of the conventional cubic unit cell, sampled at 512×512 pixels. Tilting was implemented through the multislice algorithm for large probe tilt using the method outlined in Ref. [21]. Calculations were performed using in-house code.

To identify the contribution from each atom to the ADF-STEM image, we use an effective scattering potential model to calculate the ADF STEM images [22]:

$$I(\boldsymbol{R}) = \int\int |\psi(\boldsymbol{r}_\perp, z, \boldsymbol{R})|^2 V_{\text{eff}}(\boldsymbol{r}_\perp, z) d\boldsymbol{r}_\perp dz \tag{1}$$

where ψ is the wavefunction of the elastically scattered probe electrons in the sample, calculated assuming an absorptive potential for thermal diffuse scattering, and V_{eff} is the effective scattering potential which includes information about the detector geometry. In the Einstein model for thermal scattering, V_{eff} is the sum of individual atom potentials. Though known not to be as quantitatively accurate for thicker samples as the frozen phonon model [11,12], the advantage of this model is that the ADF-STEM signal can be unambiguously decomposed to identify the individual contributions from each atom:

$$I(\boldsymbol{R}) = \sum\nolimits_i I_i(\boldsymbol{R}) \tag{2a}$$

$$I_i(\boldsymbol{R}) = \int\int |\psi(\boldsymbol{r}_\perp, z, \boldsymbol{R})|^2 V^i_{\text{eff}}(\boldsymbol{r}_\perp, z) d\boldsymbol{r}_\perp dz \tag{2b}$$

where i indexes the atoms present. Eq. (2b) identifies the contribution from scattering from each atom to the total recorded ADF-STEM signal (for each probe point). We will refer to this as the 'signal contribution'. With suitable wavefunction normalisation, Eqs. (1) and (2) give the recorded intensity (or current) as a fraction of the incident intensity (or current). All ADF-STEM signal contributions given in this paper will be expressed in these units.

3. Menu of probes

A number of different probe scenarios were simulated and compared using the criteria of interaction volume and interpretability, as defined in the introduction. Each probe is presented on its own merit, and compared to the others on the basis of possible applications.

3.1. Convergence angle

The advent of aberration correctors has enabled correction of 3rd and, in some instruments, partial correction of 5th order aberrations, minimising aberrations within a large angular range and