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Towards quantitative, atomic-resolution reconstruction of the electrostatic potential via differential phase contrast using electrons

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ABSTRACT

Differential phase contrast images in scanning transmission electron microscopy can be directly and quantitatively related to the gradient of the projected specimen potential provided that (a) the specimen can be treated as a phase object and (b) full 2D diffraction patterns as a function of probe position can be obtained. Both are challenging to achieve in atomic resolution imaging. The former is fundamentally limited by probe spreading and dynamical electron scattering, and we explore its validity domain in the context of atomic resolution differential phase contrast imaging. The latter, for which proof-of-principle experimental data sets exist, is not yet routine. We explore the extent to which more established segmented detector geometries can instead be used to reconstruct a quantitatively good approximation to the projected specimen potential.

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1. Introduction

Early work by Rose [1,2] and Dekkers and de Lang [3] shows that taking the difference between the scanning transmission electron microscopy (STEM) images from suitably configured detector segments produces images with contrast relating closely to the phase profile imparted on the electron beam by the electrostatic potential of the specimen. For a configuration involving quadrant detectors, the standard conceptual picture is that the difference signal between diametrically opposed detector segments is proportional to the beam deflection along the direction between the segments, which in turn is proportional to the gradient of the phase profile in that direction. This imaging mode is therefore called differential phase contrast (DPC). This technique has been used to great effect to image magnetic domain structure in materials [4–8], and more recently to image electric fields [9–12]. In all these cases, the lateral extent of the probe is significantly smaller than that of the variations in the magnetic or electric fields of interest – a circumstance that considerably simplifies the analysis and interpretation [13]. However, Shibata et al. [10] and Müller et al. [14] have recently demonstrated that DPC imaging can be accomplished at atomic resolution, where the probe size is larger than the scale on which the atomic potentials vary.

Fig. 1(a) shows a high-angle annular dark field (HAADF) image for a thin specimen (~30 Å) of $SrTiO_3$ viewed along the [001] zone axis, with 200 keV electrons and a probe-forming aperture semi-angle of 23 mrad. The atomic columns appear as bright peaks, with intensities roughly proportional to the square of the atomic number Z of the elements in the column, giving robustly interpretable Z-contrast imaging [15]. Fig. 1(b) shows the DPC image for quadrant detector segments aligned along the horizontal direction in the figure and spanning the scattering angle range 15.3–30.7 mrad. Each atomic column in Fig. 1(b) appears as a bow-tie pattern with one dark (negative) lobe and one light (positive) lobe. This is qualitatively consistent with the derivative in the horizontal direction of the projected electrostatic potential of the structure.[1]

A broad conceptual understanding of the appearance of the DPC image can be developed as follows. Because the fast electron penetrates the atomic electron clouds, the net force it experiences tends to be attraction to the not-fully-screened nuclei. Thus, as sketched in Fig. 1(c), when a fine electron probe passes to the left of a column of atoms it is deflected to the right by the attractive electric field of the column, increasing the signal on the right hand detector segment while reducing it on the left hand detector segment. The reverse happens when the probe passes to the right of the column. The contrast in the DPC image, which is formed from the difference between the individual signals from each detector as a function of probe position, thus reverses as the probe is scanned across an atomic column, as seen in Fig. 1(b). As recently

[1] This may be roughly gauged from the HAADF image in Fig. 1(a), which is qualitatively reminiscent of the specimen potential, save that DPC signal is also evident for the pure oxygen columns invisible in the HAADF image.

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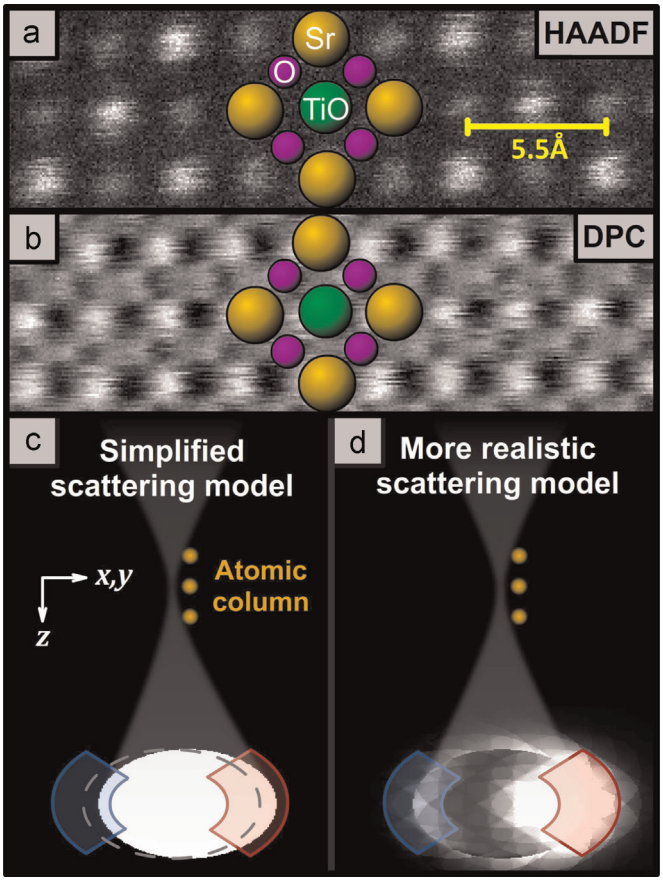

Fig. 1. Simultaneously acquired experimental (a) HAADF and (b) DPC images of $SrTiO_3$ for a 30 Å thick specimen oriented along the [001] zone axis (adapted from data in Ref. [10]). (c) Idealised DPC schematic where the STEM probe deflection results in a simple translation (from the dashed line reference) of the bright field disk across the diffraction plane and the two detector segments shown. (d) More realistic DPC schematic where the interaction of the STEM probe with the column of atoms still leads to a net deflection but the intensity redistribution in the diffraction plane is more complex.

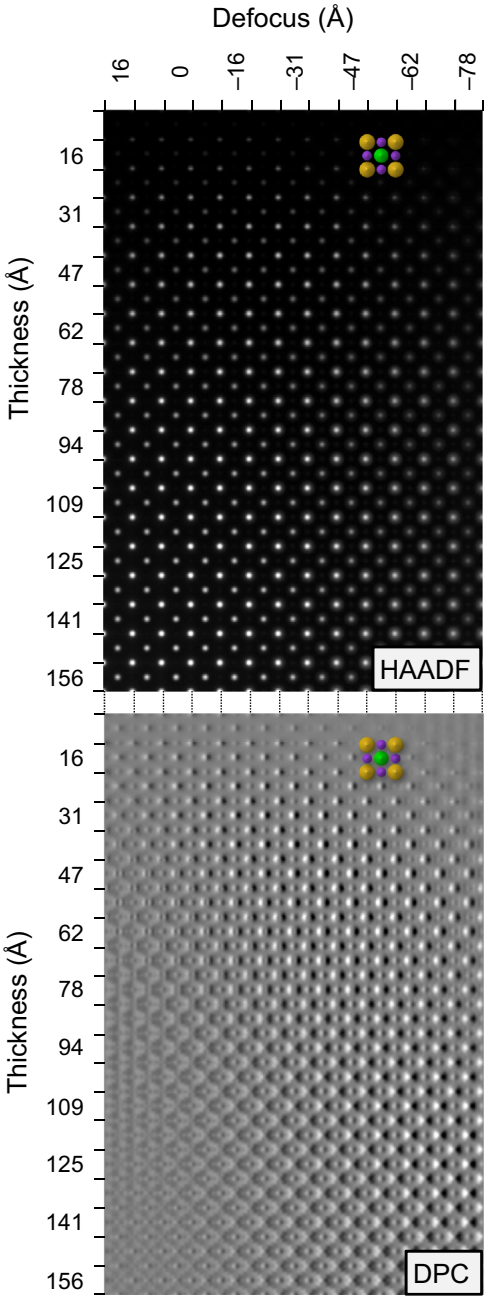

Fig. 2. Defocus-thickness tableaux for HAADF and DPC imaging of $SrTiO_3$ oriented along the [001] zone axis, assuming 200 keV electrons and a probe-forming aperture angle of 23 mrad. The HAADF detector spans the range 81–228 mrad. The DPC signal is based on diametrically opposite quadrants in a ring spanning 15.3–30.7 mrad. Positive defocus values correspond to overfocus.

emphasised by Lubk and Zweck [13] and Müller et al. [14], the "cartoon" depiction of Fig. 1(c) is rather simplistic. The reality is more like what is sketched in Fig. 1(d): due to electron scattering in the specimen, the intensity distribution in the diffraction pattern is rather more complicated than a simple rigid translation of the bright field disk. Moreover, strongly thickness-dependent electron multiple scattering and coherent interference effects in the bright field region make atomic resolution DPC imaging much less robust than HAADF. This is seen in Fig. 2 which shows defocus-thickness tableaux of HAADF and DPC images for $SrTiO_3$. Whereas the appearance of the HAADF images is largely unchanged over a wide range of thickness and defocus values, the DPC images are much more sensitive to these parameters.

The moniker "differential phase contrast" implies that the images bear close resemblance to the gradient of the phase [3,2,16,17]. However, the majority of proposals for reconstructing the specimen potential in such cases assume the specimen to be a weak phase object, and as such are not fundamentally different from other analysis/reconstruction approaches making the same assumption (e.g. Refs. [1,18–24]). In particular, Pennycook et al. have recently reconstructed the potential of a bilayer of graphene using a pixel detector [23]. However, in atomic resolution electron microscopy, the validity domain of the weak-phase-object approximation is extremely limited. It breaks down in the presence of strong scattering [25,26] and through its neglect of the spatial propagation of the wavefunction [27,28,26], especially in the high-resolution regime [26,29]. DPC imaging can to some extent overcome the strong scattering limitation: it applies equally for strong phase objects provided either that the phase gradients are constant over the size of the probe [13,30,31] or else that the displacement of the "centre of mass", i.e. the first moment, of the diffraction pattern intensity distribution can reliably be determined [13,14,30]. The neglect-of-propagation limitation, though, is at present surmountable only through comparison with detailed simulations.

In this paper we explore the validity domain of the phase-object approximation when seeking to quantitatively analyse DPC STEM imaging at atomic resolution to reconstruct the (projected) specimen potential, from which details on atom location, atom

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