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Low magnification differential phase contrast imaging of electric fields in crystals with fine electron probes

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ABSTRACT

To correlate atomistic structure with longer range electric field distribution within materials, it is necessary to use atomically fine electron probes and specimens in on-axis orientation. However, electric field mapping via low magnification differential phase contrast imaging under these conditions raises challenges: electron scattering tends to reduce the beam deflection due to the electric field strength from what simple models predict, and other effects, most notably crystal mistilt, can lead to asymmetric intensity redistribution in the diffraction pattern which is difficult to distinguish from that produced by long range electric fields. Using electron scattering simulations, we explore the effects of such factors on the reliable interpretation and measurement of electric field distributions. In addition to these limitations of principle, some limitations of practice when seeking to perform such measurements using segmented detector systems are also discussed.

1. Introduction

Determining the electromagnetic field distribution within materials is fundamental to both characterizing and understanding a variety of functional properties [1–6]. Using transmission electron microscopy we can seek to infer the field distribution by measuring the deflection of the probe electron trajectories – or, in more appropriate wave optical terms, to measure the phase shift – resulting from the Coulomb–Lorentz force from the electromagnetic fields in the sample.

A variety of imaging modes or geometries exist for imaging electromagnetic fields using the electron microscope. In conventional transmission electron microscopy, Fresnel imaging involves using large beam defocus to convert phase shifts in the exit wavefield into detectable intensity variations [7], while electron holography uses the interference between the scattered beam and a reference beam to reconstruct phase images [8–10]. In scanning transmission electron microscopy (STEM), differential phase contrast (DPC) imaging involves some measure of the deflection of the diffraction pattern as the convergent electron beam is scanned across the sample [11–13].

STEM DPC has a long history of use in imaging magnetic field distributions within materials [1–3,14,15], but has had a recent resurgence through its application to imaging electric field structure, including the piezoelectric polarization fields in quantum wells [4,16], the ferroelectric polarization fields inside ceramics [17], the spontaneous polarization in semiconductor nanowires [5], and the built-in field within a *p*–*n* junction [18]. It has also been applied to imaging the electric fields of individual atomic columns [17,19,20].

Fig. 1 shows a conceptual schematic of how STEM DPC works: the internal electric field of the sample deflects the beam across the diffraction plane. Using a position sensitive detector – Fig. 1 shows a 16 segment detector [17,21] – images can be formed in which the variation in beam deflection appears as a variation in image contrast. Fig. 1 also shows example STEM DPC images of ferroelectric $BaTiO_3$ and $LiNbO_3$ samples, formed by taking the difference between the signals from diametrically opposed detector segments. These images were obtained using an atomically fine electron probe but in a low magnification scan. Distinct contrast between regions is clearly visible in the DPC images whereas no such distinction is evident in the simultaneously acquired annular dark field (ADF) images in Fig. 1. For field mapping, the variation in contrast in the DPC images would ideally reflect the variation in strength and direction of the electric field within different domains of these ferroelectric materials. With this interpretation, Ref. [17] inferred the direction of polarization in the different domains for the $BaTiO_3$ sample. However, as we will

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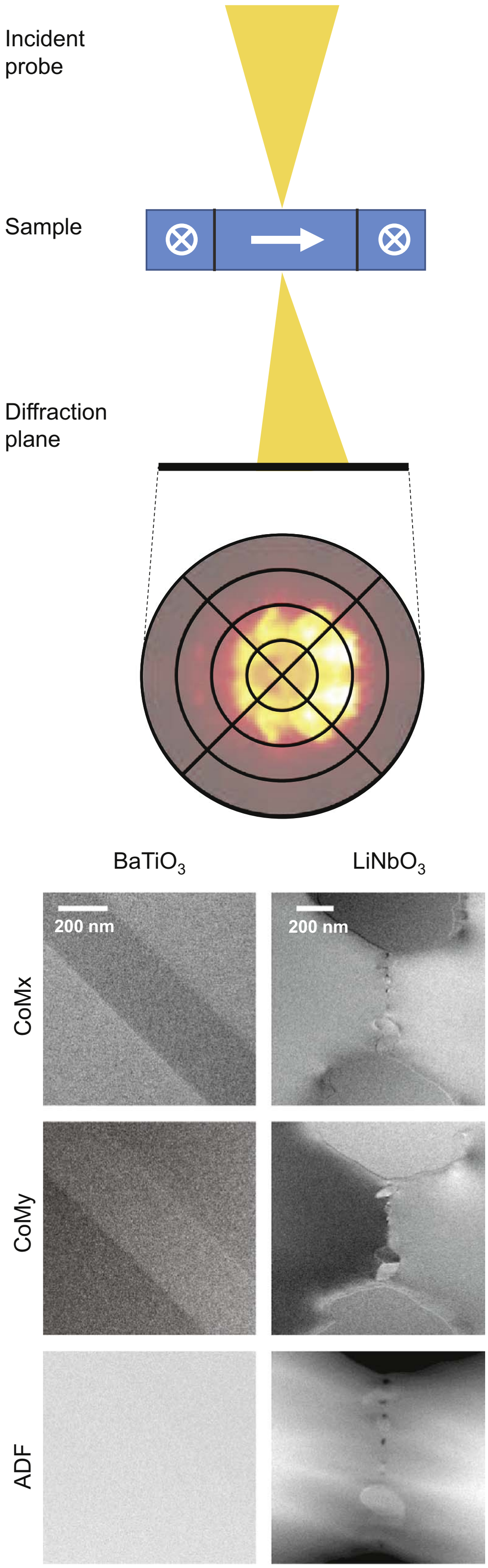

Fig. 1. Upper: simple model of beam deflection in STEM by a sample with an internal polarization (direction denoted by arrows). Lower: low magnification DPC images – calculated via centre of mass (CoM) in the horizontal x and vertical y directions – and ADF STEM images of $BaTiO_3$ and $LiNbO_3$ samples recorded using an atomically fine probe and the segmented detector pictured.

show, other factors can also contribute to DPC signals. For instance, the contrast seen in the DPC image of the $LiNbO_3$ sample in Fig. 1 may well be due to slight misorientation between grains across this fabricated grain boundary. There is hence a need to develop reliable methods for identifying and quantitatively measuring electric fields within such materials via STEM DPC.

In the absence of a sample, the diffraction pattern due to a convergent STEM probe would be a disk of uniform intensity, the so-called bright field disk. If the only interaction between the beam and sample was due to a uniform electric field in the plane of the sample, the effect on the diffraction pattern would be a simple deflection of this bright field disk, in the direction opposite to that of the field and by a magnitude related to the electric field strength via [22]

$$\theta = \frac{\lambda^2 m^* e}{h^2} E t, \tag{1}$$

where λ is the wavelength and m^* the mass of the electron, both relativistically corrected; e is the electron charge; h is Planck's constant; E is the electric field strength in the sample; and t is the sample thickness, along the beam direction, over which the field acts. A convenient metric for the bright field disk shift is the shift of the diffraction pattern's centre of mass (CoM), also known as the first moment [19], which can be measured to high precision using a pixel detector, such as CCD camera, or approximated by a segmented detector.[1]

If the phase object approximation applies, i.e. if the exit wavefield is related to the entrance wavefield via a multiplicative phase factor proportional to the projected specimen potential [23], then the electric field distribution can be determined from CoM STEM images even if the field is not uniform [19,24]. However, electrons scatter strongly from materials, and the domain of validity of the phase object approximation is thereby limited to very thin samples. For thicker crystalline samples, multiple electron scattering, also called "channelling", serves to redistribute intensity in the diffraction plane in a complex fashion, and it ceases to be clear to what extent CoM STEM images reflect the long range electric field distribution as opposed to the effects of scattering from the atomistic structure. In an extreme example, MacLaren et al. have shown that scattering from the atomistic structure at an antiphase boundary can produce a DPC signal qualitatively consistent with what one would expect from a boundary potential, even if no such potential is present [25].

In this paper we explore the effects, both qualitative and quantitative, of several key factors on the ability to accurately measure the long-range polarization of ferroelectric and polar materials via DPC STEM. We investigate the impact of channelling on the relation between the CoM signal and the assumed long-range electric field strength in bulk regions, such as domain interiors, where the long-range field is assumed to be uniform. By considering sources of noise, we explore the trade-off between samples being sufficiently thick as to give reliably detectable DPC signals and being sufficiently thin that channelling effects do not lead to ambiguous interpretation. Mechanisms which may lead to non-zero DPC signals in the absence of fields, specifically the innately asymmetric structure of polar materials and possible specimen mistilt, are also considered. Finally, we examine how the accuracy of CoM STEM images constructed using different configurations of segmented detector compares with that using pixel

[1] Although the signal in individual detector segments is not entirely linear with field, for rigid disk shift the segmented detector results need not be approximate. They can be calibrated, either experimentally using a specialized sample holder capable of applying known electric field strength [16], or through numerical modelling.