Ferroelectric Electron Holography

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Conventional TEM-investigation of ferroelectric structures has been a difficult task since decades. To assess the facilities of electron holography for the determination of the electric polarisation in ferroelectrics, a simple dipole model was set up [1]. It turns out that by interaction of the electron wave with an array of equally oriented dipoles – like in a ferroelectric domain – the electron phase is modulated in two aspects: First, each dipole modulates the wave microscopically according to the positive and negative charges in each unit cell; second, there is a mesoscopic phase shift nearly linearly increasing in direction of the polarisation (fig.1). These phase shifts arise only from the “in-plane”-components of polarisation perpendicular to the electron beam, whereas the electron wave is not affected by the “out-of-plane” components along the electron beam; Spence et al. already showed this theoretically [2]. The mesoscopic phase modulation can be computed as

$$\varphi(\vec{r}) = \frac{\sigma}{\varepsilon} \int_0^t \int \vec{P}_{ip} d\vec{r} dz$$

with $\sigma$ the interaction constant of the microscope, $\varepsilon = \varepsilon_r \varepsilon_0$ the dielectric constant, and $t$ the object thickness; $\vec{P}_{ip}$ is the in-plane component of polarisation. Assuming constant polarisation over thickness $t$, this amounts to

$$\varphi(\vec{r}) = \frac{\sigma t}{\varepsilon} \int_0^t \vec{P}_{ip} d\vec{r}$$

Consequently, the in-plane component $\vec{P}_{ip}$ can be determined from the phase image by means of

$$\vec{P}_{ip} = \frac{\varepsilon}{\sigma t} \text{grad}[\varphi(\vec{r})].$$

Using our CM200FEG-ST/Lorentz electron microscope with $\sigma = 0.0073/(V \text{ nm})$, we took holograms of BaTiO$_3$ with Lorentz-lens covering a field of view of about 800nm in square. Assuming an object thickness of $t = 50 \text{ nm}$, for BaTiO$_3$ ($\vec{P}_{ip} = \vec{P} = 26 \mu \text{C/cm}^2$, $\varepsilon_r \approx 1700$) a phase gradient of the order of $2\pi/1,000 \text{ rad/nm}$ can be expected. In the phase images, we found significant phase modulations over domains (fig.2); the surface plot of the phase image allows visual inspection in terms of polarisation; the phase gradient is more accurately displayed as an arrow plot. To determine also the out-of-plane components, one has to tilt the specimen around a well-known, arbitrary in-plane axis; from these two phase images the distribution of total polarisation $\vec{P}$ can be determined in strength and orientation.

References

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Fig. 1 Phase shift produced by a 2D-model of a ferroelectric domain made up by a mono-layer of 21*21 dipoles (left); the line scan (right) shows that - in addition to the modulation by the single dipoles - a far-reaching phase slope of the electron wave over the whole domain is found.

Fig. 2 Phase in ferroelectric domains in BaTiO$_3$ (a), phase surface (b) suggesting polarisation (c), and phase gradient as arrow plot showing polarisation distribution. No in-plane polarisation is found in the dark areas which are virtually flat.