

Model-Based Iterative Reconstruction of Charge Density in Nanoscale Materials using Off-Axis Electron Holography

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Off-axis electron holography is a powerful technique, which provides access to the phase shift of a high energy electron wave that has passed through an electron-transparent sample in the transmission electron microscope. The phase is sensitive to electromagnetic fields within and outside the specimen projected in the electron beam direction. The development of an experimental method that can be used to measure charge density distributions in materials with nanometer spatial resolution is important for understanding local material properties such as ferroelectricity, piezoelectricity and spontaneous polarization, as well as charge accumulation in ferroelectric tunnel junctions, *p-n* junctions and battery devices. Here, we discuss recent progress in the development of approaches for local charge density measurement using off-axis electron holography through the study of needle-shaped samples that were prepared for atom probe tomography. Such a charge density measurement can be used to infer the distribution of electric field around the specimen. The electric field can then be used to determine the trajectories of ions that are emitted from the needle during atom probe tomography [1].

We consider three approaches measuring charge density from phase images: i) an analytical model-dependent approach, in which a mathematical model is used to describe the charge density and phase shift; ii) a model-independent approach based on the application of a Laplacian operator to a recorded phase image [2]; iii) a model-based iterative reconstruction approach, in which a forward model is varied until a best fit to experimental measurements is obtained [3]. The analytical model-dependent approach relies on access to an analytical solution for the charge density and phase distribution for the experimental specimen geometry and requires the perturbed reference wave to be included in the model. The model-independent approach is insensitive to the presence of a perturbed reference wave and charges outside the field of view, but the measured charge density can be noisy and the result is affected by local variations in mean inner potential and specimen thickness. The model-based iterative reconstruction approach, which is described in Fig. 1, can incorporate *a priori* knowledge through the use of masks, regularization parameters and other physical constraints, resulting in lower noise but requiring care in the selection of parameters to avoid introducing artefacts. It has the further advantage that boundary pixels can be used to take account of charges outside the field of view and the perturbed reference wave. Artefacts can also be considered by assigning low confidence to regions that contain untrustworthy information. Three-dimensional charge density distributions can in principle be obtained using each approach, either by applying a backprojection-based tomographic reconstruction algorithm to projected charge density distributions measured as a function of specimen tilt angle or by using model-based iterative reconstruction. However, Fig. 2 highlights the fact that care is required to establish whether the fitted charge density is unique. It illustrates the application of model-based iterative reconstruction to a phase image of a needle-shaped sample, which contains an insulating Al₂O₃ apex that has become charged due to electron beam exposure. In this case, the fitted charge density is not unique,

as it was reconstructed only from the phase in vacuum outside the boundary of the specimen. This problem and other challenges in such measurements will be discussed.

References:

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 [4] The authors thank J Ungermann, M Riese and M Beleggia for valuable discussions.

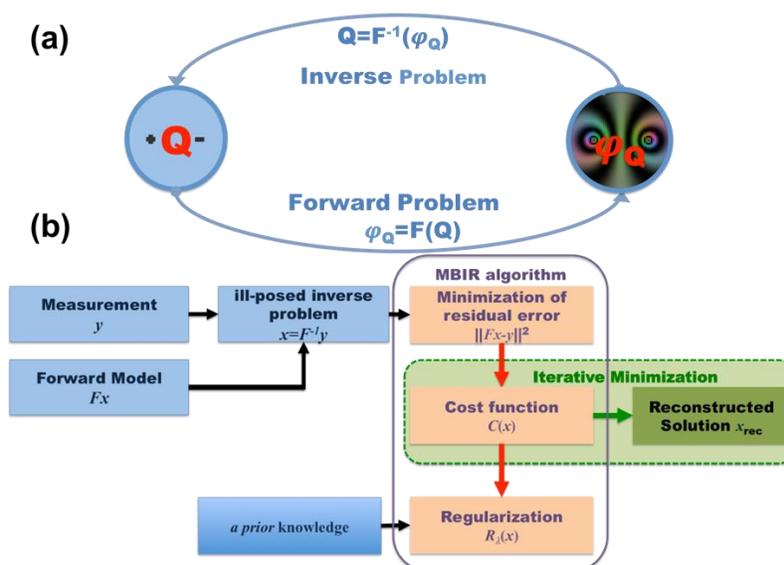


Figure 1. (a) Schematic diagram illustrating the forward and inverse problems that link the charge distribution Q in a specimen with the recorded phase shift φ_Q . (b) Workflow of the reconstruction process used to solve the ill-posed inverse problem.

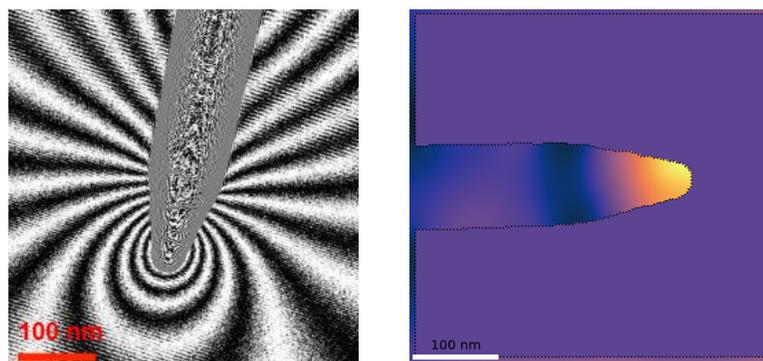


Figure 2. Left: Experimental phase contour map of an atom probe tomography needle, which has an insulating Al_2O_3 apex and a conductive base and is affected by electron-beam-induced charging. Contours inside the needle are affected by the mean inner potential and thickness profile of the specimen. The phase contour spacing is $2\pi/12$ radians. (b) Best-fitting charge density distribution in the specimen, which was reconstructed only from the phase shift outside the boundary of the needle.