# Reconstruction methods for combined HAADF-STEM and EDS tomography

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constructiebeelden, wat er toe leidt dat intensiteiten in het midden van het reconstructiebeeld onderschat worden. Figuur S2 toont een dergelijk artefact in de reconstructie van een nanodeeltje. In Hoofdstuk 5 stellen we een algoritme voor dat dergelijke niet-lineaire effecten automatisch corrigeert, zodat de HAADF-STEM data gelineariseerd kan worden. Hierna kan deze data gebruikt worden voor tomografische reconstructie op basis van HAADF-STEM + EDS.

Samenvattend wordt in dit proefschift een aantal technieken voorgesteld die een aanzienlijke verbetering kunnen geven ten opzicht van de gangbare reconstructiemethoden. De numerieke methoden kunnen toegepast worden op HAADF en EDS data die volgens gangbare technieken zijn opgenomen. Het is daarnaast mogelijk om op basis van de voorgestelde technieken de benodigde opnametijd van EDS data te verminderen. Een mogelijke toepassing van tomografie op basis van gecombineerde HAADF-STEM en EDS data, is het in beeld brengen van structuren in driedimensionale halfgeleidermaterialen. Dit maakt het onder andere mogelijk fabricagefouten te identificeren. Dit kan de verdere ontwikkeling van geavanceerde geïntegreerde schakelingen faciliteren.

# Summary

Tomography is a non-destructive technique for imaging slices (sectional images) of an object. The slices are computed from a series of projection images taken from different angles. This computation is known as tomographic reconstruction. The principle of tomography can be applied to different techniques for acquiring the projection images. For instance, the widely-used computed tomography (CT) for medical diagnostics is based on X-ray imaging. Standard X-ray CT can only resolve structures that are at least about one hundred micrometers in size. In the life sciences and materials science, the size of critical structures can sometimes be as small as a few atoms. For instance, the size of structures in computer chips is nowadays around 10 nanometers. In these fields, the principle of tomography is combined with electron microscopy (EM), a combination which is known as electron tomography (ET). EM uses accelerated electrons for imaging and can resolve structures at the atomic scale.

The research in this thesis is focused on tomographic reconstruction based on two imaging modalities in EM. The first modality is *high angle annular dark field scanning transmission microscopy* (HAADF-STEM), and the second modality is *energy-dispersive X-ray spectroscopy* (EDS). Figure S1 shows examples of projection images acquired by HAADF-STEM and EDS. While HAADF-STEM is a standard modality in EM nowadays, EDS is sometimes used as a supplementary technique to resolve more chemical information. HAADF-STEM yields a single-channel image that mixes the information for all chemical elements. Using

a standard imaging scheme, HAADF-STEM images have relatively low noise levels. EDS, on the other hand, yields multiple element-specific images. However, the resulting projection images have much higher noise levels. Tomography based on EDS is more challenging compared to HAADF-STEM, due to the strong Poisson noise, the limited number of projection angles and the long data acquisition time.

Considering the complementary properties of the two modalities, in Chapter 2 we propose an approach to perform element-specific reconstructions from HAADF-STEM and EDS tomographic data that are simultaneously acquired. Using this technique, it is possible to obtain element-specific reconstructions with better image quality compared to only using EDS data.

In this approach, element-specific projection images are required for all chemical elements present in the sample, which may be not possible in practice due to limitations of the EDS imaging technique. In Chapter 3, we propose a different approach for combining HAADF-STEM and EDS, which improves the element-specific reconstruction from EDS data by encouraging the reconstructed images to have common edge locations with the reconstruction from HAADF-STEM data.

A key problem with incorporating such prior knowledge is to know which method works well on which type of sample. In Chapter 4 we present a framework for constructing advanced reconstruction methods as a "recipe" that is tailored to the particular sample. We present guidelines for determining which method should be used depending on the experimental conditions and sample properties.

Applying ET on samples with large thicknesses is challenging due to the dominant nonlinear effects in HAADF-STEM data. As a consequence, cupping artifacts can appear in the reconstructed tomographic images, which means that the image intensities being underestimated in the center of reconstruction. Figure S2 shows the cupping artifact present in the reconstructed image for a nanoparticle. In Chapter 5, we propose an algorithm for correcting these nonlinear effects automatically, so that the HAADF-STEM data can be linearized and subsequently used for HAADF-STEM + EDS tomographic reconstruction.

Overall, several approaches are given in this thesis for improving the image quality of element-specific tomographic reconstructions. The numerical methods can be applied to HAADF and EDS data acquired in a conventional manner using the existing electron microscopic technique. It is also possible to design novel data acquisition-processing pipelines based on them, to reduce the time for EDS data acquisition. A potential application of HAADF-STEM + EDS tomography is imaging structures and faults in 3D semiconductor materials, which can facilitate the development of advanced integrated circuits.