

3D Imaging of Nanomaterials by Discrete Tomography

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Abstract:

The field of discrete tomography focuses on the reconstruction of samples that consist of only a few different materials. Ideally, a 3D reconstruction of such a sample should contain only one grey level for each of the compositions in the sample. By exploiting this property in the reconstruction algorithm, either the quality of the reconstruction can be improved significantly, or the number of required projection images can be reduced. The discrete reconstruction typically contains fewer artifacts and does not have to be segmented, as it already contains one grey level for each composition.

Recently, a new algorithm, called DART, has been proposed that can be used effectively on experimental electron tomography datasets. In this paper, we propose discrete tomography as a general reconstruction method for electron tomography in materials science. We describe the basic principles of DART and show that it can be applied successfully to three different types of samples, consisting of embedded ErSi₂ nanocrystals, a carbon nanotube grown from a catalyst particle, and a single gold nanoparticle, respectively.

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1. Introduction

Electron tomography is a powerful tool for investigating the three-dimensional (3D) morphology and inner structure of nanomaterials [1-7]. Single electron microscopy images are inherently two-dimensional (2D), which limits their use for studying the 3D structure of materials. In electron tomography, projection images of the sample are acquired from a range of different angles by tilting the sample relative to the electron beam. Using a reconstruction algorithm that can combine the information from the 2D projection images, a full 3D reconstruction can be obtained. Imaging modes that can be used for tomography include BF TEM [7], HAADF STEM [1], and ADF TEM [5]. At present, most reconstructions are computed using either backprojection schemes or iterative reconstruction algorithms, such as SIRT [8-11]. The Weighted Backprojection algorithm has been used extensively for electron tomography of biological samples since the early 1980s (reviewed in [12]). More recently, iterative methods have become popular, mainly due to a major increase in the available computation resources [13].

All of these reconstruction methods require a large number of projection images to obtain results of acceptable quality. Preferably, more than 100 projections are used, tilting the sample in steps of 1-2 degrees over a range of at least $\pm 60^\circ$ [11, 14]. Using fewer projections results in more severe reconstruction artifacts. Even if many projections are available, the reconstruction often contains artifacts due to the limited tilt range during acquisition. Such artifacts are known as *missing wedge artifacts*. The effect of the missing wedge can be reduced in one direction by acquiring projection data using two perpendicular tilt-axes [15,16]. This technique reduces the missing wedge to a “missing pyramid”. The resulting reconstructions are more accurate compared to single-axis tomography, but residual artifacts due to the missing angles remain. Furthermore, dual-axis tomography significantly increases the complexity of the acquisition and alignment procedures.

After the reconstruction has been computed, the 3D volume has to be interpreted to obtain meaningful information about the sample. This can be done either visually or, in some cases, automatically. To obtain quantitative results about morphology and composition of the sample, the reconstruction must be segmented: the voxels of the reconstructed volume must be separated into different classes, corresponding to the different compositions in the sample. Due to the various artifacts in the reconstruction, segmenting tomographic images can be very hard; labor intensive manual segmentation is often the only option and even then it can be very difficult to clearly identify the boundaries between different materials. Moreover, any manual segmentation will always be biased in some way.

The field of discrete tomography focuses on the reconstruction of samples that consist of only a few different materials [17, 18]. Ideally, a reconstruction of such a sample should contain only one grey level for each type of material in the sample. By exploiting this property in the reconstruction algorithm, the quality of the reconstruction can be improved and, in some cases, the required number of projection images can be reduced substantially. Two rather different variants of discrete tomography have been proposed in the literature.

The first variant can be applied to the reconstruction of nanocrystals at atomic resolution [19]. In that case, two forms of “discreteness” are exploited simultaneously: first, it is assumed that the crystal contains only a few types of atoms (i.e., image intensities). Second, it is also assumed that the atoms lie on a regular grid, of which the general structure is known in advance. Although these assumptions are quite strict, they are also powerful, as they allow for the reconstruction of

three-dimensional images from very few projection images recorded along crystallographic zone axes. Potential advantages of this technique include the reconstruction of defect structures such as vacancies.

Another variant of discrete tomography, which we cover in this paper, can be applied at lower resolution, where individual atoms cannot be resolved. In this approach, it is assumed that the reconstruction contains only a few different intensities corresponding to a limited number of well defined materials, whereas no assumptions are made on a possible lattice structure of the sample. Although the mathematical theory behind discrete tomography has been studied since the 1990s, the technique has not yet been used in many practical applications, due to the fact that no efficient and robust algorithm was available. Recently, a new algorithm for discrete intensity tomography, called DART (Discrete Algebraic Reconstruction Technique), was proposed in [20] that is capable of reconstructing large 2D images (i.e., 1024x1024) in a few minutes on a standard PC. The same approach can be used for reconstructing 3D volumes, treating the volume as a stack of 2D slices. To the authors' knowledge, DART is the first algorithm that can be used on images of this size, making it practical to use discrete intensity tomography for electron tomography.

In this paper, we propose discrete tomography, and DART in particular, as a general tool for electron tomography of materials science samples. The basic principles of the DART algorithm are described in Section 2.

In Section 3, we apply DART to three different experimental electron microscopy datasets. The first sample consists of ErSi_2 nanoparticles embedded in SiC. The results for this dataset show that DART yields accurate results in reconstructing homogeneous nanoparticles. The second sample is a catalyst particle used to grow a bamboo-like carbon nanotube. The particle consists of both Cu and Cu_2O . Reconstruction of the catalyst particle provides an illustration of the capabilities of DART for more complex structures that have several compositions, corresponding to more than two grey levels in the reconstruction. The third sample consists of a single gold nanoparticle, of which only 15 projections were recorded. The results for this dataset show that DART is capable of reconstructing accurate reconstructions from very few projection images, contrary to continuous methods. For each of the three cases, we describe simulation experiments that demonstrate how the DART algorithm performs in a theoretical, optimal scenario. Subsequently, we show the reconstruction results obtained by applying DART to experimental electron tomography data.

Section 4 provides an outlook on the use of discrete intensity tomography as a general tool for materials science applications. Conclusions will be made in Section 5.

2. The DART algorithm

In this section, the basic steps of the DART algorithm will be reviewed briefly.

DART is an iterative algorithm that combines a continuous iterative reconstruction algorithm (such as ART, SIRT) with discretization steps. In this paper, SIRT is used as the underlying continuous reconstruction algorithm (see Chapter 7 of [21]). We recall that SIRT is an iterative reconstruction algorithm, that computes an approximate solution of the linear system $\mathbf{Ax} = \mathbf{b}$, where the vector $\mathbf{x}$ contains the grey level for each pixel, the vector $\mathbf{b}$ contains the measured

projection data, and the matrix $\mathbf{A}$ describes the projection process (i.e., computing the product $\mathbf{Ax}$ yields the projections corresponding to the image $\mathbf{x}$). If no exact solution of this system exists, SIRT computes a solution for which the norm of the difference $\|\mathbf{Ax} - \mathbf{b}\|$ (called *projection error*) between the computed projection and the measured data is minimal w.r.t. a certain vector norm, i.e., a least-square solution (see [22] for details).

As an example, suppose that we want to reconstruct two nanoparticles that have a single composition, embedded in a homogeneous support. Figure 1a shows a cross-section of the sample, orthogonal to the rotation axis of the tilt stage. We assume that the projection data are available for a tilt range from -60 to +60 degrees, using angular steps of 10 degrees (i.e., 13 projections).

Before applying DART, one needs an estimate of the *number* of grey levels in the reconstruction (i.e., the number of materials in the sample), as well as the *actual grey levels*. Such an estimate can be hard to obtain, in particular when only a small number of projections are available. As a first step before applying DART, a conventional SIRT reconstruction is usually computed to obtain information about the materials in the sample and their grey levels. A SIRT reconstruction of the simulated nanoparticles is shown in Fig. 1b). Based on this SIRT reconstruction, and possibly additional knowledge about how the sample was created, we now assume that the particles consist of a single material and, consequently, two grey levels should be used for the reconstruction. Estimates of the actual grey levels to be used in the reconstruction can be obtained by selecting one or more regions that are clearly contained within the particles (or the background) and averaging the grey level in the SIRT reconstruction for those regions.

Although the nanoparticles have a constant density, the SIRT reconstruction exhibits a spectrum of grey values. In addition, the shapes of the reconstructed particles are clearly distorted, due to the missing wedge and the small number of projections used. The range of grey levels in the reconstruction immediately presents a problem when the reconstructed image needs to be segmented. Segmentation is commonly performed by thresholding, but it is not obvious at all which threshold should be chosen in this case. The DART algorithm starts from a thresholded SIRT reconstruction, and then iteratively improves upon the current segmentation. Although a threshold also has to be chosen for DART, its choice is of low importance to the final result. In this example, we choose the threshold to be exactly in the middle between the grey level of the background and the particle in the original image.

Figure 1c shows the thresholded SIRT reconstruction. The thresholded reconstruction shows that pixels of the interior of the object that are not too close to the boundary are assigned the correct segmentation class (either interior or exterior). Pixels that are close to the boundary can be detected automatically from the thresholded SIRT reconstruction, by checking if any of the surrounding pixels belong to a different segmentation class. We refer to these pixels as *boundary pixels*, and to the remaining pixels as *non-boundary pixels*. Figure 1d shows the boundary of the two particles that was computed from Fig. 1c. Note that even some non-boundary pixels have the wrong grey level in the thresholded SIRT reconstruction, when compared to the original phantom image. For example, this occurs in the region at the top of the image, between the two particles.

We now turn back to the SIRT reconstruction in Fig. 1b. The non-boundary pixels in the interior of the particles are assigned the grey level that corresponds to the nanoparticles, whereas the non-boundary pixels in the background are assigned the grey level that corresponds to the background (typically 0). Next, the SIRT algorithm is used again, but only the boundary pixels are allowed to vary. The non-boundary pixels are kept fixed at their discrete levels. In this way, the number of variables in the linear equation system $\mathbf{Ax}=\mathbf{b}$ is significantly reduced, while the number of equations remains the same. Figure 1e shows the result after 10 SIRT iterations for the boundary pixels. Note that the grey levels for the particle and the background have been scaled, in order to display the darker and brighter grey levels that appear in the reconstructed boundary.

The red arrow in Fig. 1e indicates a region of the new SIRT reconstruction that is even darker than the background. This is an indication that near this region, pixels have been erroneously assigned the particle grey level (i.e., a high grey level), instead of being classified as background pixels. To compensate for this error, the nearby boundary pixels will assume a very low grey level. The boundary that results from the new set of SIRT iterations can be quite rough, as all pixels are allowed to vary independently. A smoothing step is performed on the boundary to partially remove this roughness. The result is shown in Fig. 1f. Applying a stronger smoothness filter leads to less noise and smoother boundaries in the reconstruction, while losing some fine single-pixel details. In all experiments in this paper, a Gaussian blur is applied with a radius of 1 pixel.

The image in Fig. 1f can be considered as a continuous reconstruction, just as the SIRT reconstruction in Fig. 1b. We now repeat the same steps as before, starting from the reconstruction in Fig. 1f. The result of the threshold step is shown in Fig. 1g. It is already very clear that the quality of the segmentation has improved considerably in just one iteration. Fig. 1h shows the reconstruction result after 20 iterations, which is nearly perfect w.r.t. the original phantom. Figure 2 shows a flow chart of the basic steps performed in the DART algorithm.

3. Three case studies

In this section, we report on electron tomography experiments that have been carried out for three different samples. For each of the samples, we first describe its composition and the acquisition conditions. An important obstacle towards quantitative evaluation of the reconstruction quality is the lack of “ground truth”, obtained by alternative methods. For electron tomography, alternative methods that can be used for comparison are often not available. To estimate the quality difference between the continuous SIRT reconstruction and the discrete DART reconstruction, a simulation experiment was performed for each of the samples. Based on simulated projection data computed from a phantom image, SIRT and DART reconstructions are computed and subsequently compared to the original phantom. Although our simple simulation experiments cannot replace a real experiment, they still provide useful insights in the quality difference that can be expected between SIRT and DART. For each of the samples, the simulation experiments are followed by a visual comparison between SIRT and DART for the real experimental dataset.

In all experiments, both for simulated and experimental data, the SIRT reconstruction has been computed using our own SIRT implementation based on Chapter 7 of [21]. The same SIRT implementation is also used for computing the start solution in DART, and for each of the

intermediate continuous DART steps. The fact that the same SIRT implementation was used in both algorithms makes it easier to compare the results, as implementation details are the same for both cases. All SIRT reconstructions have been obtained after 25 SIRT iterations. Typically, the quality of the SIRT reconstruction does not improve further by performing more iterations. The SIRT reconstruction was subsequently used as the initial reconstruction for DART, which was run for 50 iterations. Note that a DART iteration typically takes about half the time of a SIRT iteration, as many pixels are kept fixed and do not have to be updated.

The first sample consists of ErSi_2 nanoparticles embedded in SiC. It is assumed that both the nanoparticles and the substrate can be represented by constant grey levels in the reconstruction, yielding a binary reconstruction problem. The second sample is a catalyst particle, which consists of two different compositions and has several cavities. This case study demonstrates the capabilities of DART for more complex structures that have several compositions. The third sample consists of a pentagonal shaped gold nanoparticle. The experimental dataset contains only 15 projections, which results in low reconstruction accuracy for continuous reconstruction methods. The results for this dataset demonstrate that DART can compute highly accurate reconstructions even from very few projection images.

Case study I: ErSi_2 nanocrystals

Sample description

ErSi_2 nanocrystals have been prepared in SiC by high-dose ($10^{16}/\text{cm}^2$) and high-temperature (700C) ion implantation of Er in SiC followed by rapid thermal annealing (1600C) [23]. In this manner, nanoparticles with a diameter of just a few atoms to about 25 nm are formed in a narrow band underneath the SiC surface. Furthermore, small voids and partially filled voids are formed close to the surface during the rapid thermal annealing. In 2D, the structure and shape of these particles has been extensively studied [24-27] and the shape of the particles was described as a hill-like structure; see Fig. 3. In order to better understand the shape of these particles, we used electron tomography to evaluate the full 3D structure of these nanocrystals [28].

The tilt-series for the tomographic reconstruction was acquired in HAADF-STEM mode (inner angle of detector = 70 mrad) on a Tecnai F20 ST with a nominal spot size of 0.2 nm. Using the Xplore3D package, 177 images were acquired semi-automatically over a tilt-range of $\pm 77^\circ$. The images were aligned using a combined cross-correlation and marker tracking approach in IMOD, which allowed for the correction of slight image rotation, magnification, and tilt-angle changes.

Phantom study

Simulation experiments were performed in order to assess the reliability of the DART reconstruction, as well as the quality difference between SIRT and DART that can be expected in a real experiment. The simulation experiments were based on a single 2D slice, which resembles a cross-section through the SiC sample (Fig. 4a). Simulated projections were computed using the same tilt angles used in the real experiment. To make the simulation more realistic, Gaussian noise was added to each of the projection images and each of the projection images was mis-

aligned by a small, random amount of at most one detector pixel in either direction. The resulting projections are used as input for a SIRT (Fig. 4b) and DART (Fig. 4d) reconstruction.

The effect of the missing wedge can be clearly seen in the streaking artifacts. A major advantage of using DART is that it allows one to quantify the observations in a straightforward way since the reconstruction is already segmented. This is in contrast with SIRT reconstructions that often have to be segmented in a manual and therefore subjective way. Figure 4c shows the SIRT reconstruction after thresholding. Although different thresholds can be used to get more accurate results in various parts of the image, a single threshold that gives optimal results throughout the image cannot be found. The DART reconstruction shows that even small details are reconstructed accurately and that there are no apparent missing wedge artifacts.

Reconstruction results for the experimental dataset

The experimental HAADF-STEM dataset was reconstructed using SIRT and DART. For computing the DART reconstruction, it was assumed that the reconstruction should contain only two grey levels, for the particles and the background. The grey level of the particles is required as an input parameter for DART. The appropriate grey level was determined based on the SIRT reconstruction, by measuring the grey level in the inner region of several large particles and averaging the result. Figure 5 shows a comparison between reconstructed slices for SIRT and DART in the xz plane (viewing in the direction of the tilt axis) and xy plane (viewing in the direction of the 0° beam). When viewing in the direction of the tilt axis, reconstruction artifacts due to the missing wedge or misalignment of the projection data are usually most visible. The reconstruction results appear to correspond quite well with the simulation results from Figure 4. From the SIRT reconstructions, one can see with reasonable accuracy what the shape of each of the particles is. The particles at the extreme left and right part of the reconstruction have far worse quality, as they fall outside of the field-of-view in many of the projection images and, consequently, have a much larger missing wedge of projection angles (and therefore would never be used in a regular continuous reconstruction). Even these particles appear to be reconstructed well by DART.

Figure 6 provides a comparison between segmentations obtained by SIRT and DART, for a different slice in the xz-direction. Fig. 6c-e show thresholded SIRT reconstructions, where the threshold has been set at varying levels. As the larger particles are brighter in the SIRT reconstruction than the smaller ones, it is not possible to find a single threshold such that the segmentation result is satisfactory for all particles simultaneously. Obtaining an accurate segmentation of the SIRT reconstruction, including all the particles of different sizes, still requires hand-work. In the DART reconstruction, it appears that the resulting segmentation contains nearly all particles, except possibly some extremely small ones and some non stoichiometric features with a reduced Er to Si ratio. The results for this dataset illustrate that DART can significantly improve the reconstruction quality for binary images, in particular if a segmented reconstruction is required for further quantitative analysis.

Case study II: catalyst particle in a carbon nanotube

Sample description

Carbon nanotubes have been grown by chemical vapor deposition starting from alkali-element modified Cu/MgO catalysts. We refer to [28] for more details on the growth process of the sample. It was found in [6] that the catalyst particle consists of both Cu and Cu₂O. The resulting carbon nanotubes have a so-called bamboo-like structure. The catalyst material is not only present at the tip of the nanotube, but is also partially filling the hollow bamboo compartments within the nanotube.

The tilt-series of a single carbon nanotube (including the catalyst particle) for the tomographic reconstruction was acquired in HAADF-STEM mode (inner angle of detector = 70mrad) on a Titan 80-300 microscope (FEI) using a Fischione ultra-high-tilt tomography holder. A series of 66 images was acquired automatically over a tilt range of -54 to $+74^\circ$, with projections taken every 2 degrees. FEI Inspec3D™ was used to align the 2D projections of the HAADF-STEM tilt series by cross correlation. Figure 7 shows a single projection image from the tilt-series.

Phantom study

Simulation experiments were performed to compare the reconstruction using SIRT and DART with a known phantom image. The simulation experiments were based on a single 2D slice, which resembles a cross-section through the catalyst particle; see Fig. 8a. Simulated projections were computed using the same projections as for the real experiment. To make the simulation more realistic, Gaussian noise was added to each of the projection images and each of the projection images was misaligned by a small, random amount of at most one detector pixel in either direction. The resulting projections are used as input for a new SIRT and DART reconstruction. Fig. 8b shows the resulting SIRT reconstruction. The effect of the missing wedge can be clearly seen in the streaking artifacts. The DART reconstruction is shown in Fig. 8c. It corresponds very well with the original phantom image. Moreover, the DART reconstruction is already segmented: each pixel is assigned to either the background, the Cu or the Cu₂O compositions.

Reconstruction results for the experimental dataset

A reconstruction of the sample was computed from the measured experimental data, using both SIRT and DART. For the DART reconstruction, three grey levels were used corresponding to the background, Cu and Cu₂O. The assumption that three grey level should be used was made after studying the SIRT reconstruction. The grey levels for both compositions were estimated from the continuous SIRT reconstruction by averaging the intensity over a group of voxels that clearly contain a single composition.

Fig. 9 shows three orthogonal cross-sections through the reconstructed volumes, for both SIRT and DART. Missing wedge artifacts are clearly visible in the xz slice of the SIRT reconstruction

(vertical elongation/smearing) as well as in the yz slice (horizontal elongation/smearing). The boundary regions between the background and the catalyst particle and between both compositions contain a wide range of grey levels, visible as a blurring of the boundary. This makes it very difficult to properly determine which voxels belong to the background, Cu, or Cu₂O. In the DART reconstruction, the position of the boundary has already been determined by the reconstruction algorithm. Moreover, similar to the results of the simulation experiments, the DART reconstruction does not seem to suffer from the typical missing wedge artifacts (elongation and smearing) present in the continuous reconstruction. The DART reconstruction is suitable for further automatic morphological analysis, and therefore allows for the extraction of all sorts of quantitative information about the catalyst, such as the ratio of both compositions, the shape and size of the cavities in the catalyst, etc.

The results for this dataset illustrate that DART can also be used if the sample contains several compositions, corresponding to more than two grey levels in the reconstruction, as long as the compositions are separated spatially.

Case study III: gold nanoparticle

Gold nanoparticles were synthesized in a multi-step process. First, a solution of small gold nanoparticle seeds (solution A) was obtained by the addition of 0.1 mL of a 0.01M NaBH₄ solution to 5ml of a 0.1M CTAB and 1.25×10^{-4} M HAuCl₄ solution. Then, 0.5 ml of solution A was added to 5 ml of a growth solution (B) (0.1 M CTAB, 2.5×10^{-4} M ascorbic acid and 1.25×10^{-4} M HAuCl₄). The seeds of solution A are allowed to react for several minutes in solution B, producing a colloidal solution (C) of larger anisotropic Au nanoparticles. Then, 0.5 ml of solution C was added to 5 ml of the growth solution B and left to react for 30 minutes. The final solution contains anisotropic Au nanoparticles, as revealed by the HAADF-STEM images seen in the experimental tilt series (see Fig. 10).

The tilt-series was acquired in HAADF-STEM mode (inner angle of detector = 35 mrad) on a Tecnai F20 microscope (FEI) using a Fischione ultra-high-tilt tomography holder. A tilt series of 15 images was acquired manually from -48° to $+74^\circ$ with projections recorded at the following angles: -48° , -46° , -40° , -30° , -20° , -10° , 0° , $+10^\circ$, $+20^\circ$, $+30^\circ$, $+40^\circ$, $+50^\circ$, $+60^\circ$, $+70^\circ$ and $+74^\circ$. FEI Inspect3D™ was used to align the 2D projections of the HAADF-STEM tilt series by cross-correlation. The gold particles are deposited on an amorphous carbon support. The (weak) contrast of the support was subtracted from the projection data in a preprocessing step, resulting in a reconstruction problem for two grey levels (gold and “background”) for DART.

Phantom study

Based on a SIRT reconstruction of the experimental dataset (see Fig. 12), we designed a phantom image that resembles a slice through the experimental dataset (see Fig. 11a). It contains two grey levels, for background and gold. Simulation experiments were performed for the phantom image, using both SIRT and DART to compute a reconstruction from 15 projections, using the same tilt angles as for the experimental dataset. Figure 11b shows the resulting SIRT reconstruction.

Reconstruction results for the experimental dataset

Reconstructions have been computed from the experimental dataset using both SIRT and DART. Compared to the previous two case studies (I: ErSi_2 nano particles and II: carbon nanotube with catalyst particle), the number of projection images is much smaller. This poses a severe problem for continuous reconstruction algorithms.

The DART reconstruction was computed using two grey levels, for the gold and the background. Determining the grey level for the interior of the gold particle was a trial-and-error procedure. Setting the grey level too high resulted in holes in the interior of the particle, as well as an irregular boundary. We gradually decreased the grey level until the resulting reconstruction had a rather smooth boundary and no holes. We admit that this manual procedure is unsatisfactory and are currently looking into computational techniques that allow for estimation of the grey level as part of the reconstruction algorithm.

Reconstruction results for slices in the xz, xy and yz directions are shown in Fig. 12 for both SIRT and DART. The DART reconstruction is much sharper than the SIRT reconstruction and does not show any missing wedge artifacts, as we already expected from the simulation experiment. Moreover, the SIRT results for the experimental dataset closely resemble the experimental reconstruction, which confirms the validity of the simulation experiment.

In Fig. 13 we show a 3D surface rendering of the DART and SIRT reconstructions. The SIRT reconstruction clearly suffers heavily from missing wedge artifacts and an insufficient number of projection images, whereas the DART reconstruction corresponds closely with the regular pentagonal shape. All facets of the particle are clearly visible in the DART reconstruction. Note that the visualization of the SIRT reconstruction can be changed substantially by setting a different threshold for the surface, but doing so does not yield a visual improvement.

4. Discussion

The three experimental datasets have been recorded using different microscopes with varying imaging conditions and by different microscopists. The three specimens have greatly varying material characteristics. Using discrete intensity tomography for these datasets did not require any adjustments of the acquisition procedure. In fact, the decision to attempt a reconstruction using discrete tomography was made *after* acquisition in all three cases. This clearly shows that discrete intensity tomography can potentially be applied to a wide range of materials samples.

The experimental tilt-series of the ErSi₂ nanoparticles contains images for high tilt-angles ($\pm 77^\circ$), which limits the negative effects of the missing wedge. From the continuous SIRT reconstructions, it is already possible to determine the location and shape of each nanoparticle with reasonable accuracy. However, automatic segmentation of the SIRT reconstruction is not straightforward. When a single global threshold is used to separate the particles from the background, either small details are no longer visible (threshold set too high) or the size of some reconstructed particles becomes larger than they appear to be in the original reconstruction. Careful manual segmentation can alleviate these problems to some extent, but is time consuming and prone to bias. The DART reconstruction shows all features of interest in a single segmented image, which is immediately ready for further quantitative processing (i.e., volume determination, morphological analysis). We believe that DART is a very powerful approach to reconstructing homogeneous nanoparticles in general.

The second experimental dataset, containing a catalyst particle in a carbon nanotube is more complex: three grey levels have to be used in the reconstruction, for the background, Cu and Cu₂O. The morphology of the sample is also more challenging, as the catalyst particle contains holes. As the tilt range for this dataset is significantly smaller compared to the ErSi₂ dataset (-54 to $+74^\circ$), the effects of the missing wedge are more pronounced in the SIRT reconstruction. Our simulation experiments show that DART can potentially remove these effects completely. The reconstruction results for the experimental dataset seem to match very well with the simulation experiment. The DART reconstruction provides a segmented volume, where each voxel is assigned to either the background, the Cu or the Cu₂O composition.

In the third dataset, containing a pentagonal shaped gold nanoparticle, the SIRT reconstruction clearly suffers heavily from missing wedge artifacts and a sparse data set (of only 15 projections). The optical properties of noble metal nanoparticles are strongly dependent on their size and morphology and small changes to their physical shape can lead to significant changes in their far-field optical properties (extinction, scattering and absorption spectra) as well in the near-field (evanescent fields and local field enhancement properties). For accurate metrology (surface area, volume, angles between facets, etc), the results shown in this third case show that DART is capable of reconstructing accurate reconstructions from very few projection images.

Discrete tomography provides a method for reducing missing wedge artifacts, as well as other types of artifacts, without the need to improve image acquisition. DART reconstructions typically contain less noise and the boundaries between the different compositions are clearly visible. The technique is particularly powerful if further morphological processing is to be performed after the reconstruction, as the discrete reconstruction is already segmented. An important restriction for the use of discrete intensity tomography is that the set of intensities must be known *before* the reconstruction. In many cases, the grey levels can be estimated from the SIRT reconstruction and subsequently be used in DART. This approach was followed for the first two experimental datasets described in this paper. If the SIRT reconstruction is of very poor quality and contains many artifacts (such as for the gold sample), there is no clear procedure for determining the grey levels. In future research, we aim at automatic estimation of the grey levels as part of the reconstruction algorithm.

5. Conclusions

In this paper, we have presented a new reconstruction technique for electron tomography of materials science samples. Discrete tomography can be used if the sample consists of only a few different compositions, each corresponding to a constant grey level in the reconstruction. Although the mathematical foundation of discrete tomography has been well established for years, its use for electron tomography has been hampered by the lack of an efficient reconstruction algorithm. The DART algorithm has a running time comparable (up to a factor of about three) with the running time of the SIRT algorithm for continuous tomography. In this paper, we demonstrated that DART can yield improved reconstruction quality compared to SIRT for a wide range of experimental electron tomography datasets. Compared to continuous tomography, DART reduces the effects of the missing wedge and experimental noise. A major benefit is also that the reconstructed volume has already been segmented, so that it can be used directly for further quantitative processing.

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Figure captions

- Figure 1. Illustration of the main steps of DART. a) phantom image of two nanoparticles; b) SIRT reconstruction; c) thresholded SIRT reconstruction; d-g) intermediate results of various steps of DART; h) final DART result.
- Figure 2. Flow chart of the DART algorithm
- Figure 3. a) HAADF-STEM image showing the size distribution and shape of the ErSi₂ nanocrystals; b) HAADF-STEM image showing a projection of the atomic structure of a hill-like nanocrystal (published with permission from [26]).
- Figure 4. a) phantom image, resembling a slice through the SiC layer. b) SIRT reconstruction from 177 projections over a tilt-range of $\pm 77^\circ$. c) DART reconstruction. d) thresholded SIRT reconstruction.
- Figure 5. SIRT and DART reconstruction of the experimental dataset for a slice in the xz-plane and a slice in the xy-plane.
- Figure 6. Comparison between DART and SIRT for the experimental dataset, where the SIRT reconstruction has been thresholded at varying greylevels.
- Figure 7. HAADF-STEM image from the acquired tilt series showing the catalyst particle and the bamboo-like carbon nanotube.
- Figure 8. a) Simulated 2D slice through the top part of the catalyst. b) SIRT reconstruction from simulated projections between -54° and $+74^\circ$, where noise and a small misalignment has been applied to each projection image. c) DART reconstruction from the same projection data as the SIRT reconstruction in b).
- Figure 9. Top: Three orthogonal slices through the SIRT reconstruction in the a) xz-plane b) xy-plane and c) yz-plane. Bottom: Corresponding slices through the DART reconstruction (def).
- Figure 10. HAADF-STEM image from the gold nanoparticle experimental tilt series.
- Figure 11. a) Phantom, resembling a partial slice of the sample; b) SIRT reconstruction based on the same 15 projection angles as the experimental dataset c) DART reconstruction from the same projections.
- Figure 12. SIRT and DART reconstruction of the experimental gold dataset
- Figure 13. 3D volume rendering of the SIRT and DART reconstructions of the gold nanoparticle. Figures a) and b) show two different views of the SIRT

reconstruction. Figures c) and d) show the corresponding views of the DART reconstruction.

Figure 1.

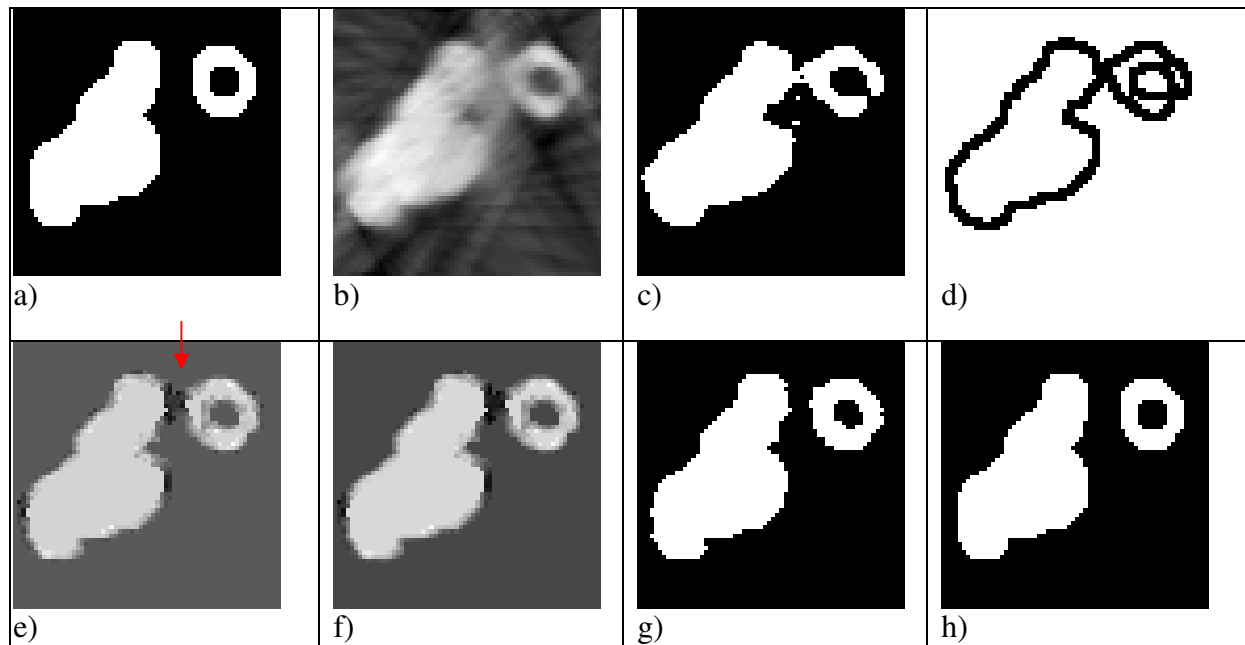


Figure 2.

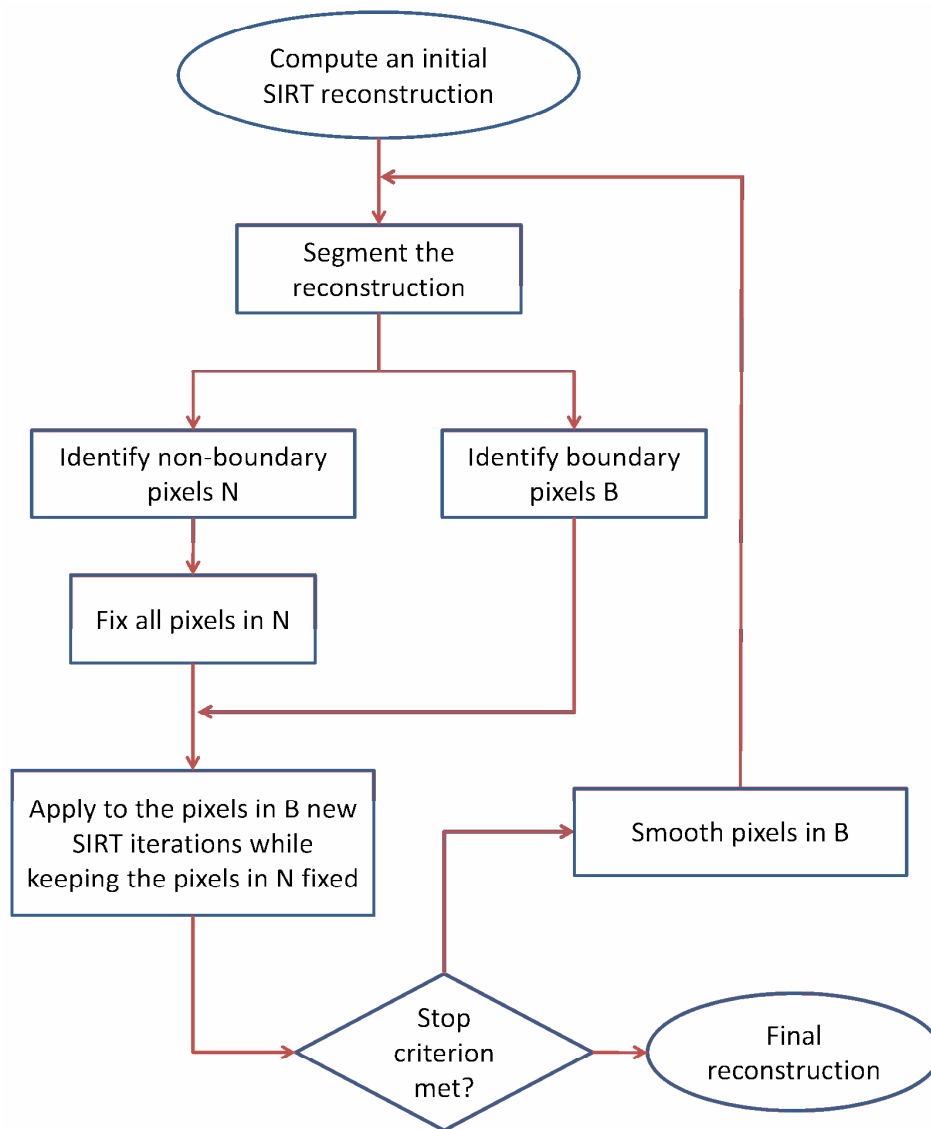


Figure 3.

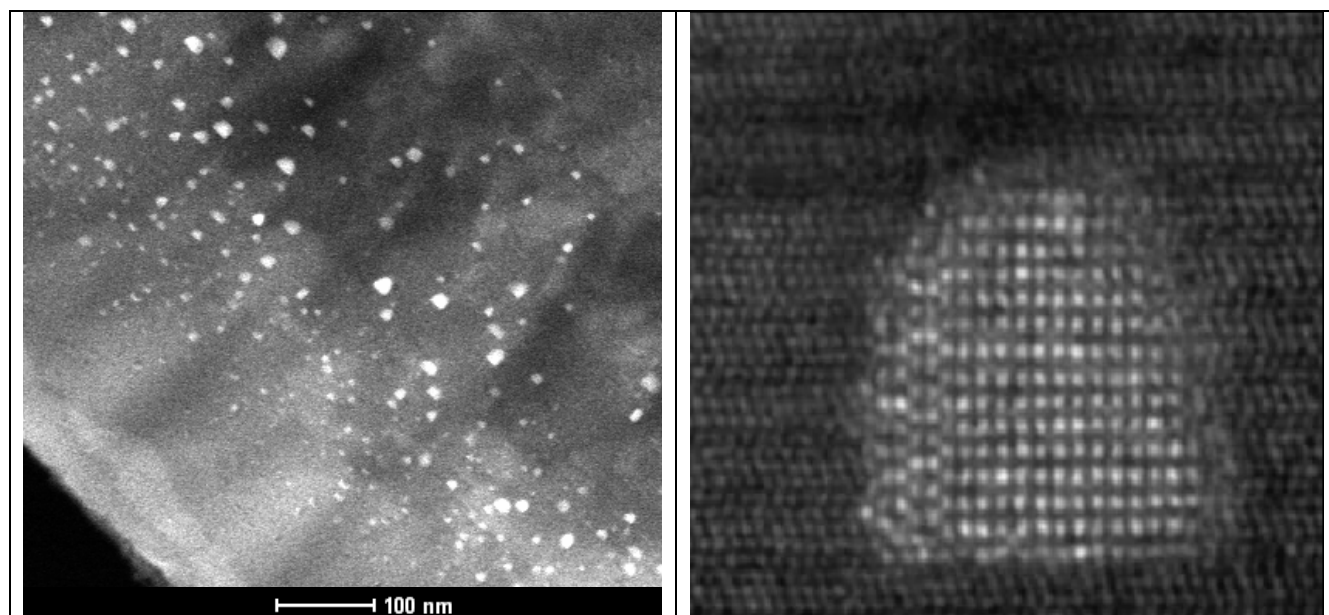


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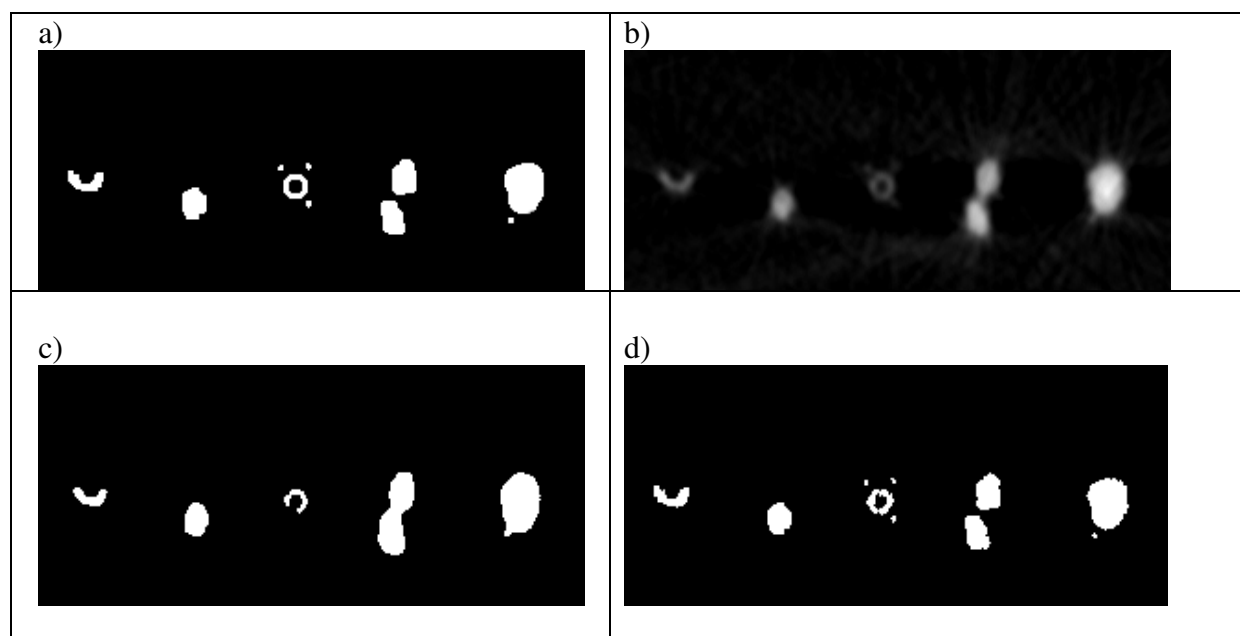
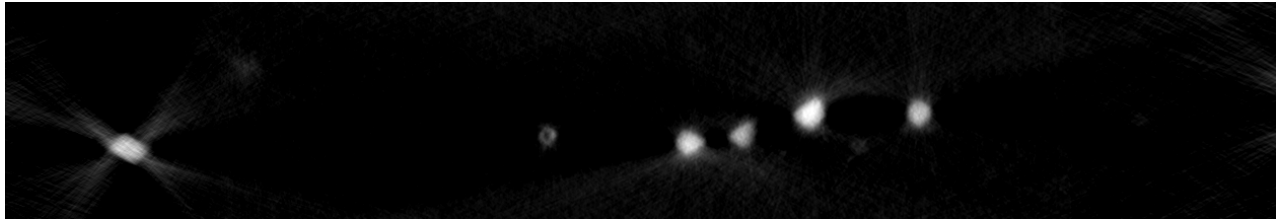


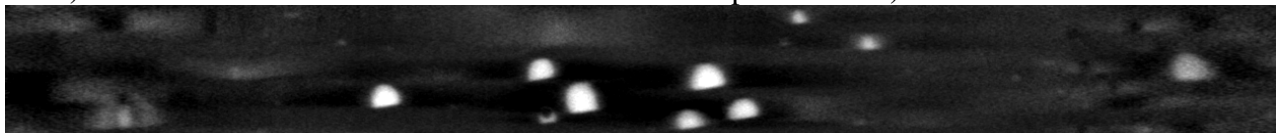
Figure 5.



a) SIRT reconstruction of a slice in the xz-plane



b) DART reconstruction of the same slice in the xz-plane as in a)

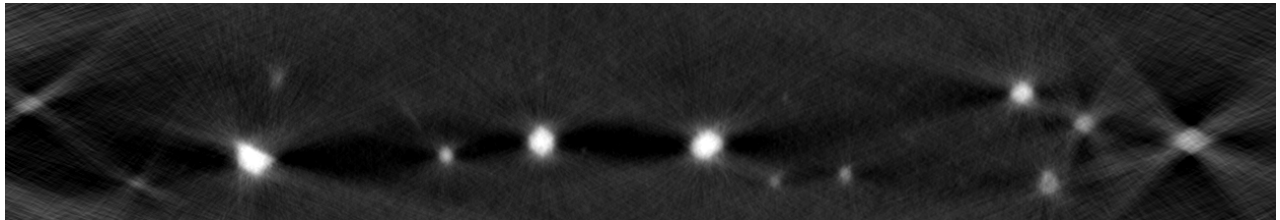


c) SIRT reconstruction of a slice in the xy-plane



d) DART reconstruction of the same slice in the xy-plane as in c)

Figure 6.



a) SIRT reconstruction of a slice in the xz-plane



b) DART reconstruction of the same slice in the xz-plane as in a)



c) SIRT reconstruction after thresholding: low threshold



d) SIRT reconstruction after thresholding: medium threshold



e) SIRT reconstruction after thresholding: high threshold

Figure 7.

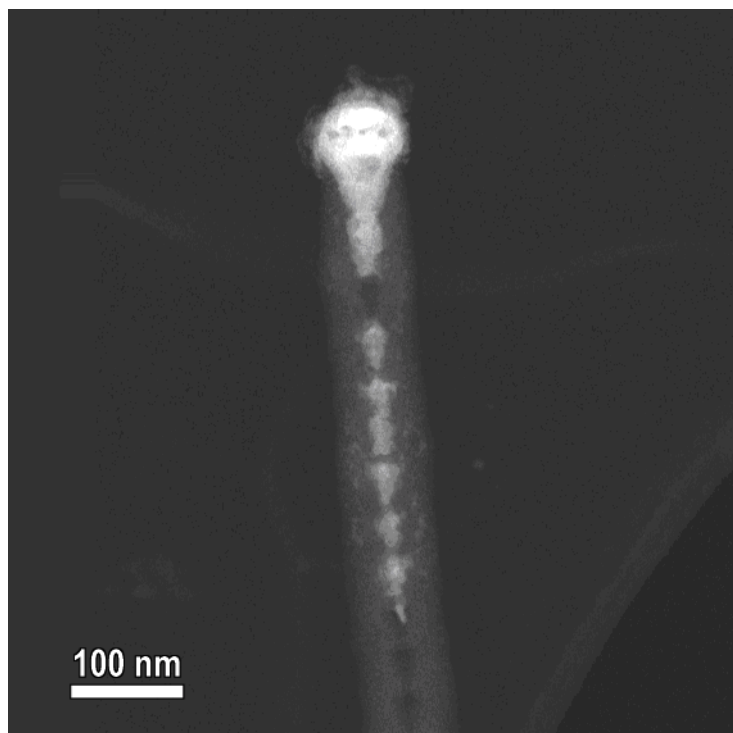


Figure 8.

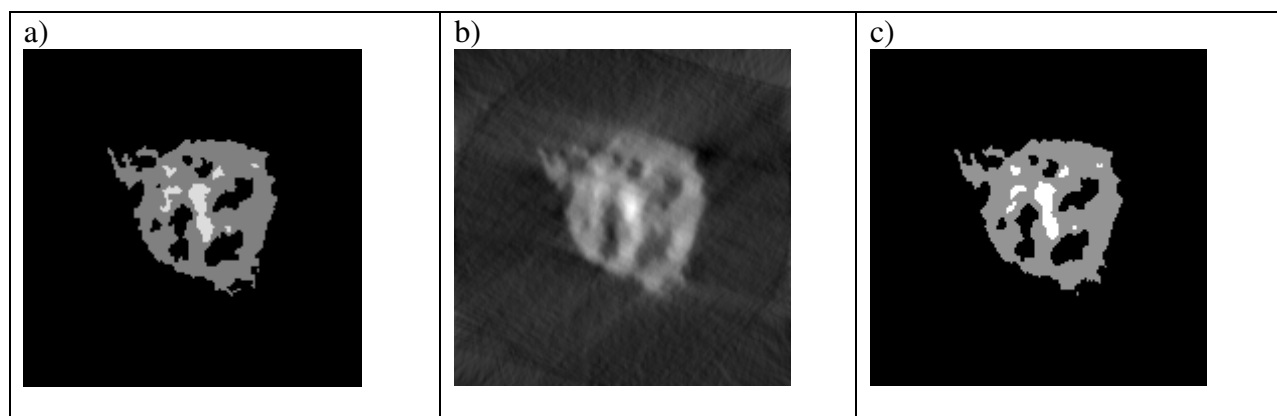


Figure 9.

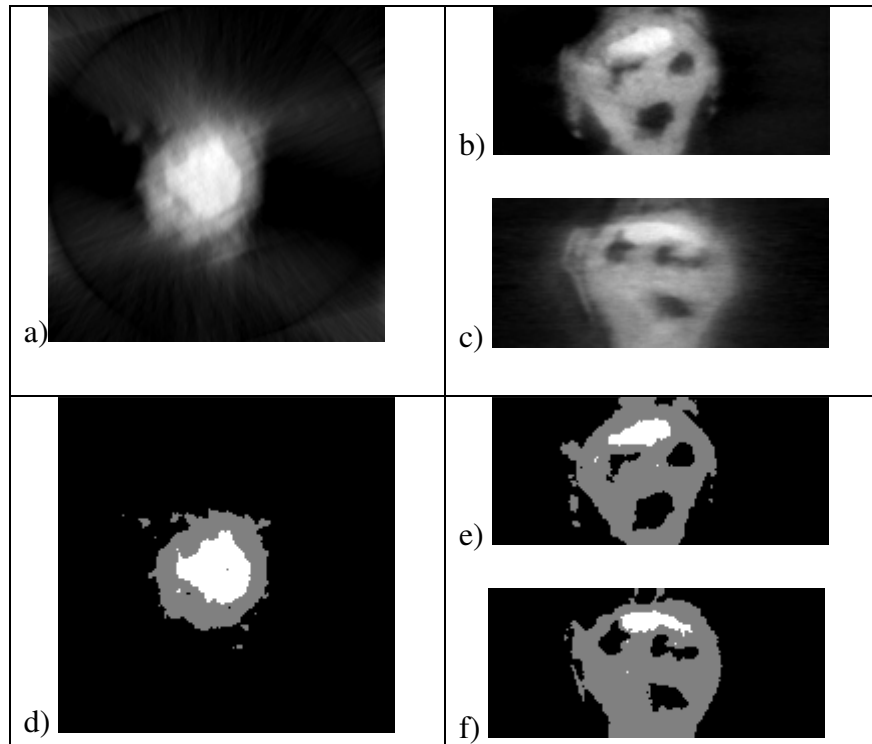


Figure 10.

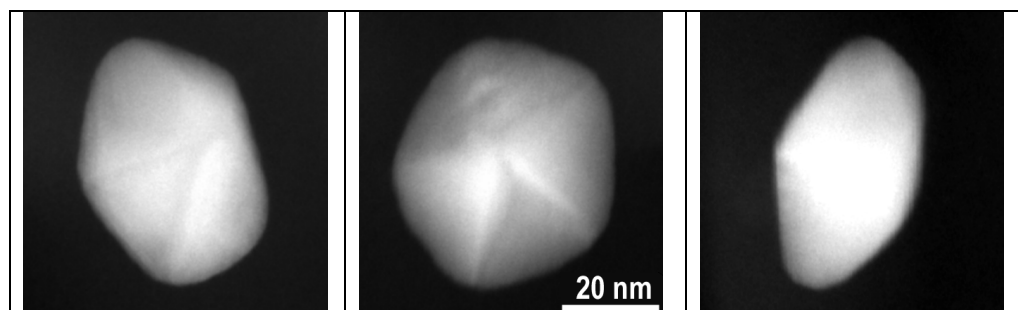


Figure 11.

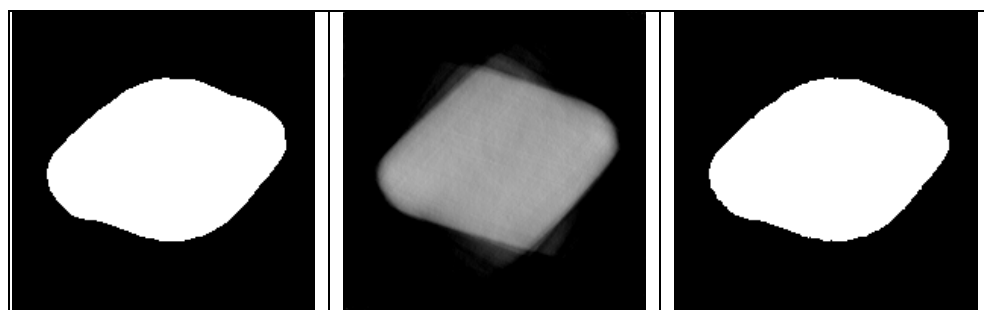
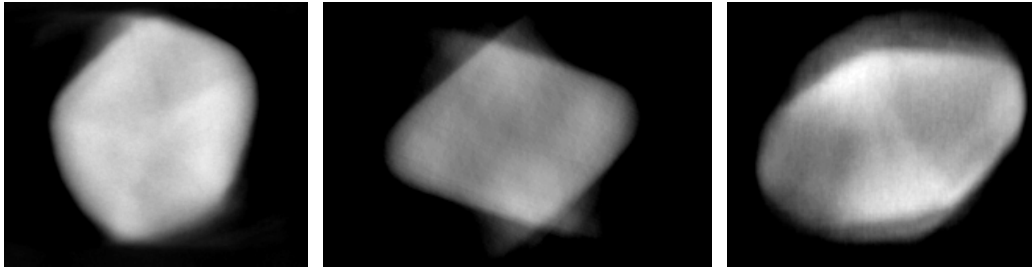


Figure 12.

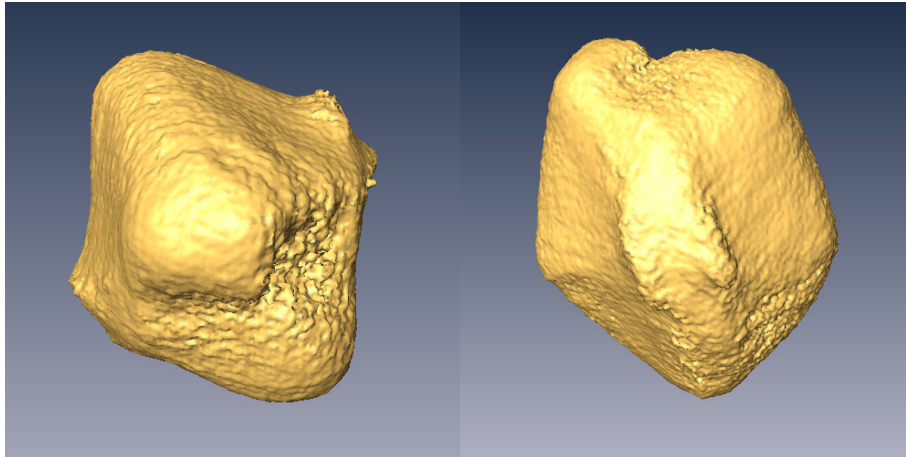


a) Cross-sections through the experimental SIRT reconstruction in the xy-plane, xz-plane and yz-plane



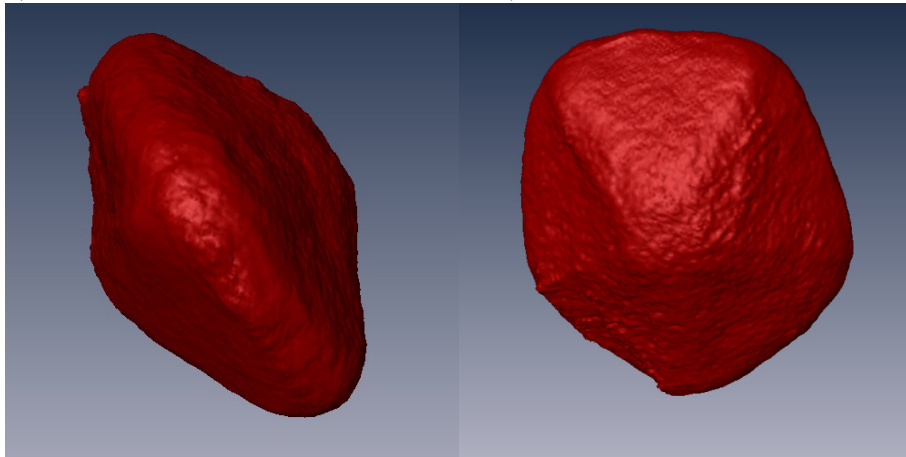
b) Cross-sections through the experimental DART reconstruction in the xy-plane, xz-plane and yz-plane

Figure 13.



a)

b)



c)

d)