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Electron tomography based on highly limited data using a neural network reconstruction technique

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ABSTRACT

Gold nanoparticles are studied extensively due to their unique optical and catalytical properties. Their exact shape determines the properties and thereby the possible applications. Electron tomography is therefore often used to examine the three-dimensional (3D) shape of nanoparticles. However, since the acquisition of the experimental tilt series and the 3D reconstructions are very time consuming, it is difficult to obtain statistical results concerning the 3D shape of nanoparticles. Here, we propose a new approach for electron tomography that is based on artificial neural networks. The use of a new reconstruction approach enables us to reduce the number of projection images with a factor of 5 or more. The decrease in acquisition time of the tilt series and use of an efficient reconstruction algorithm allows us to examine a large amount of nanoparticles in order to retrieve statistical results concerning the 3D shape.

1. Introduction

Gold nanoparticles (NPs) have truly unique electronic, optical as well as catalytic properties, rendering them ideal for numerous applications in fields as diverse as photovoltaics, optoelectronics and biomedicine [1–4]. Furthermore, gold NPs can be prepared with almost any desired shape. Crucial to their application, however, is their exact structure, and specifically their anisotropy as well as the surface facets they expose. Currently, it is empirically understood how particle size and shape may be controlled during synthesis [5–8]. Although transmission electron microscopy (TEM) has become a routine tool to investigate e.g. particle size, (atomic) structure and shape, increasingly advanced TEM is required for a more in-depth characterisation. For example, the surface facets of Au nanorods have a major influence on crucial effects such as reactivity and ligand adsorption and there has been controversy regarding facet indexing [9–11]. Indeed, TEM images are only two-dimensional (2D) projections of three-dimensional (3D) objects. To overcome this problem, 3D electron microscopy, or “electron tomography” was developed [12,13]. In 2003, Paul Midgley and co-workers demonstrated the potential of the technique in materials science based on high angle annular dark field scanning transmission electron (HAADF-STEM) microscopy [14,15]. Since then, different electron microscopy modes have been combined successfully with tomography, leading to a broad variety of 3D structural and compositional information at the nanoscale [16–21]. Very often, electron tomography is used to determine the size and shape of the particles and nowadays, 3D reconstructions can even be obtained with a resolution at the atomic level [22,23]. Although these investigations provide very precise information on the NP morphology, both the acquisition of tilt series as well as the 3D reconstruction is very time consuming and it is consequently not straightforward to acquire results in 3D that are statistically relevant, which is a major drawback e.g. when using electron tomography to optimize the synthesis of NPs. This problem will be even more essential for anisotropic NPs that are currently receiving a lot of attention because of the increased flexibility they provide to tune the final (optical) properties [24–26]. Since the optimization of the production of NPs with a specific shape would largely benefit from statistical 3D results with a nanometer resolution, one of the emerging challenges in the field of electron tomography is to increase the throughput of 3D reconstructions of NPs. At the same time, the quality of the reconstructions should be maintained and should enable one to obtain reliable and quantitative results concerning parameters such as particle size and surface morphology.

In this paper, we will determine the 3D shape and size of a

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large set of anisotropic Au NPs. We will make effective use of a new approach for electron tomographic reconstructions that is based on artificial neural networks. The neural network filtered backprojection method (NN-FBP) is a recently developed reconstruction technique that has been applied successfully to X-ray tomography [27]; however the implementation for electron tomography is completely new. The method that we propose will enable us to reduce the number of necessary projection images for a 3D reconstruction by a factor of 5 or more. In this manner, the acquisition time and time that is necessary for a 3D reconstruction is significantly reduced, enabling 3D results that are of statistical relevance.

2. Neural network filtered backprojection method

The sample that was investigated contains Au NPs yielding different morphologies: nanorods, nanotriangles, nanoprisms and nanospheres. An HAADF-STEM overview image of the sample is provided in Fig. 1a. Although this image only corresponds to a 2D projection of a set of 3D objects, it is already clear that different morphologies occur. In conventional electron tomography, a large set of 2D projection images is acquired from the same region of interest over a large tilt range with a tilt increment of typically 1° or 2°. As all the investigated nanoparticles have a thickness below 100 nm, the projection requirement for tomography is satisfied [14,28]. Once this so-called "tilt series" is aligned, the images serve as an input for a mathematical algorithm that enables one to reconstruct the original 3D structure. Very often, the 3D reconstruction is performed using the "Weighted Backprojection" algorithm (also known as Filtered Backprojection) or using the "Simultaneously Iterative Reconstruction Technique" (SIRT). The outcome of this procedure for the different NPs in Fig. 1a is visualized in Fig. 1b. The reconstructions are calculated using the SIRT algorithm and are based on a series of 151 images, acquired over a tilt range of $\pm 75°$. Since the quality of 3D reconstructions based on the conventional approach is predominantly determined by the number of projection images [29–31], these experiments are very time-consuming and require sufficient measurement time at the TEM.

The key to increasing the image quality if only a small number of 2D projections are available, is the effective use of prior knowledge in the reconstruction. By exploiting rather generic features of the particles, without assuming a specific shape or morphology, this additional knowledge is used to compute a particle shape that better approximates the true morphology. Various algorithms involving prior knowledge are currently in use in electron tomography (e.g. the DART algorithm for discrete tomography [32] and multiple methods for Total Variation Minimization [33]), where the particular prior knowledge is encoded by the user and various parameters have to be set. These prior-knowledge based methods are typically very time-consuming, which limits the throughput of 3D reconstructions that can be achieved by using them for reconstruction. Furthermore, implementing these methods can be difficult and time-consuming as well, since they rely on advanced mathematics. In this paper, we propose an alternative approach called Neural Network Filtered Backprojection (NN-FBP) that was first described in [27], which can effectively exploit sample characteristics to improve reconstruction quality, while still being highly computationally efficient. Here, we apply this new technique for the first time to electron tomography data. The application of NN-FBP to electron tomography consists of two phases: (i) a *learning phase*, in which full tilt series and their corresponding reconstructions are used to calibrate the reconstruction algorithm and (ii) a *reconstruction phase*, in which large batches of limited tilt series (i.e. using fewer projections) are rapidly reconstructed. A schematic overview the NN-FBP method is given in Fig. 2. In the next subsections, we will first briefly explain how the reconstructions are formed in the reconstruction phase, followed by an overview of how the calibration is performed in the learning phase.

2.1. Reconstruction phase

Reconstructions obtained by standard Weighted Backprojection are commonly plagued by a range of reconstruction artefacts when reconstructing from a limited tilt range and few projection angles. Streaks can be observed due to the limited number of projections, and the limited angular range leads to elongation and blurring in the *Z*-direction. In [27], it was found that strong improvements on

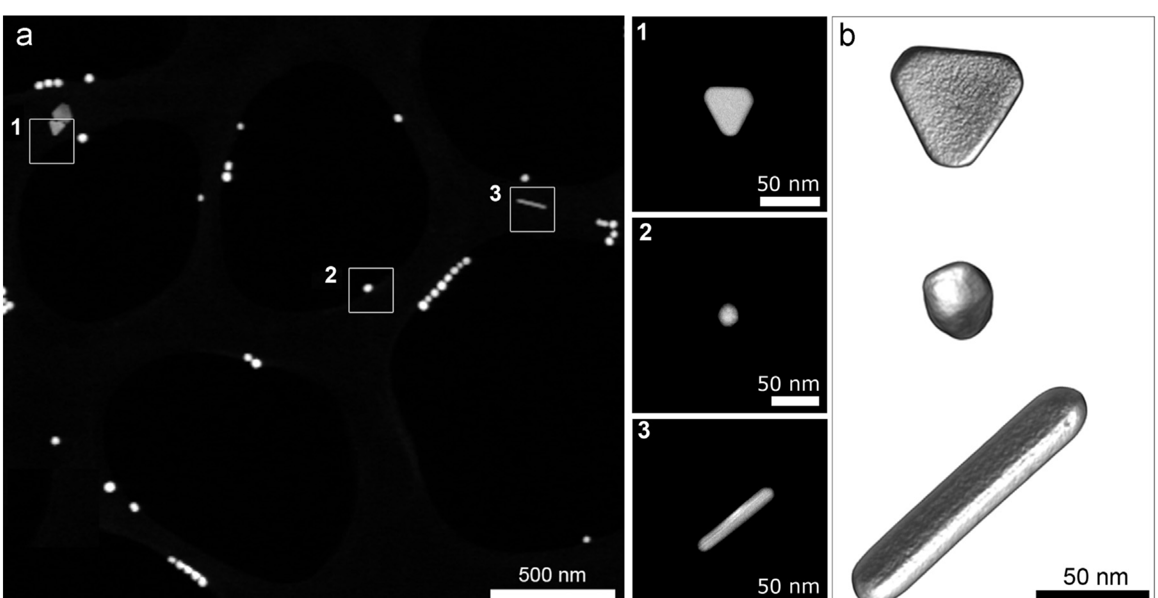

Fig. 1. (a) The HAADF-STEM overview image shows the presence of several morphologies in the sample, with indication of (1) a nanotriangle, (2) a nanosphere and (3) a nanorod. (b) 3D volume renderings of the corresponding nanoparticles are presented.