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Throughput maximization of particle radius measurements through balancing size versus current of the electron probe

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ABSTRACT

In this paper we investigate which probe size maximizes the throughput when measuring the radius of nanoparticles in high angle annular dark field scanning transmission electron microscopy (HAADF STEM). The size and the corresponding current of the electron probe determine the precision of the estimate of a particle's radius. Maximizing throughput means that a maximum number of particles should be imaged within a given time frame, so that a prespecified precision is attained. We show that Bayesian statistical experimental design is a very useful approach to determine the optimal probe size using a certain amount of prior knowledge about the sample. The dependence of the optimal probe size on the detector geometry and the diameter, variability and atomic number of the particles is investigated. An expression for the optimal probe size in the absence of any kind of prior knowledge about the specimen is derived as well.

1. Introduction

In this paper we investigate the optimal probe settings for high angle annular dark field scanning transmission electron microscopy (HAADF STEM). It is common practice to optimize the resolution of the coherent probe contribution in some respect, see for example Ref. [1] for a derivation of the Scherzer settings or Ref. [2] for a derivation of the settings when the limiting aberrations are of higher order. In these studies the incoherent probe contribution due to a finite source size is ignored, assuming a purely coherent point source. However, a point source emits no current, so that no electrons would be present for the actual imaging. Introducing a finite source size broadens the probe in a way that is well parameterized by Barth and Kruit in Ref. [3]. The optimal probe size will depend on a trade-off between the probe width and the beam current. A large width increases the beam current and augments the signal-to-noise ratio, although at the expense of reduced resolution. In practice, operators balance these two effects by adjusting the so-called spot size of the microscope. However, this choice may be somewhat subjective and therefore operator dependent. In this paper we provide a sound theoretical basis for this choice. The problem we investigate, is maximizing the throughput when measuring the radii R of spherical nanoparticles deposited on a uniform support. That is, given a prespecified precision of the estimates of R, we seek the probe size that yields the minimum required recording time needed to reach that precision.

The images are considered as data planes from which structural information has to be estimated quantitatively. For this we use a model for the object and for the imaging process, including electron–object interaction, microscope transfer and image detection. This model describes the expectations of the intensity observations and it contains the parameters that have to be measured. These parameters are determined by fitting the model to the experimental data by the use of a criterion of goodness-of-fit, such as least squares or maximum likelihood. In this way structure determination becomes a statistical parameter estimation problem. The precision with which structure parameters can be estimated is limited by the presence of noise. Use of the Fisher information [4] allows to derive an expression for the best attainable precision with which the structure parameters can be estimated. This expression, which is called the Cramér–Rao lower bound (CRLB), is a function of the object parameters, the microscope parameters and the electron dose.

Statistical optimal design is a discipline that if applied to electron microscopy, searches the set of microscope parameters that yields the highest attainable precision on the estimates of one or several of the structure parameters of the sample. In Ref. [5] for example, the CRLB on the variance with which atom column positions can be estimated is used as a performance measure in the optimization of STEM experiments. This methodology has been applied to optimize the design of other microscopy experiments as well, see Refs. [6–9] for examples. In this article, a lower bound σ^2_{CR} on the variance of the estimate of R is derived. In our current problem, σ^2_{CR} is a function of R, the parameter that has to be

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estimated and thus is unknown a priori. This problem is common for any optimal experimental design model involving a non-linear statistical model [10]. For this reason, we adopt a Bayesian approach in which we use a prior probability distribution, $p(R)$, which reflects the distribution of R and seek the probe size that is optimal over the entire distribution of radii. In order to derive an overall optimal probe size, we define σ_{B}^2 as the average $\sigma_{\mathrm{CR}}^2(R)$ weighted by $p(R)$. It is explained in Section 4 that the probe size optimizing σ_{B}^2 also accomplishes maximum throughput. Therefore, in the remainder of this paper we will look for the probe optimizing σ_{B}^2 with constant recording time per unit area whilst keeping in mind that this same probe also maximizes throughput.

The outline of this paper is as follows. In Section 2, we introduce the models for the probe, the particles and the support and combine them into a model for the images. In Section 3, we specify the joint probability function, and explain how it leads to the Fisher information matrix, and the CRLB. In Section 4, we introduce the Bayesian optimality criterion, show the equivalence between minimum σ_{B}^2 and maximum throughput, give analytical results and rules of thumb for the optimal probe sizes, and carry out a simulation study to check the influence of the particles' atomic numbers, the detector geometry, and the mean and variance of the prior distribution $p(R)$. In Section 5, we summarize the final conclusions.

2. The image model

2.1. Probe model

In Ref. [3], Barth and Kruit propose a root-power-sum algorithm that relates the probe current I to the probe size d_p, where d_p is the diameter of the disc containing p% of the total probe current. In this paper, we choose d_{50} as a resolution measure, as suggested in Refs. [3,11].

The dependence of d_{50} on the microscope settings is given by

$$d_{50}^2=(d_I^{1.3}+(d_A^4+d_s^4)^{1.3/4})^{2/1.3}+d_c^2 \tag{1}$$

with

$$d_I=\frac{2}{\pi}\frac{1}{\alpha}\sqrt{\frac{I}{B_rE_0}},$$

$$d_A=0.54\frac{\lambda}{\alpha},$$

$$d_s=0.18C_s\alpha^3$$

and

$$d_c=0.34C_c\frac{\delta E}{E_0}\alpha.$$

In these expressions, B_r is the reduced brightness [12] of the electron gun, E_0 is the acceleration voltage, α is the semi-angle of the aperture selecting the spot size, λ the electron wavelength, C_s the spherical aberration, C_c the chromatic aberration, and δE the full width at half the maximum of the electrons' energy distribution.

Eq. (1) can be used to produce $(d_{50},I)-$curves by fixing I at different values and minimizing d_{50} numerically with respect to α for each of the I values. The solid curve in Fig. 1 was calculated in this way, with the microscope parameters used throughout this paper and given in Table 1. This approach is somewhat unwieldy. Therefore, we will derive an analytical, albeit approximate, expression for the $(d_{50},I)-$curves, providing more insight in the problem at hand, that is, the maximization of throughput of particle radius measurements through optimization of the probe size d_{50}.

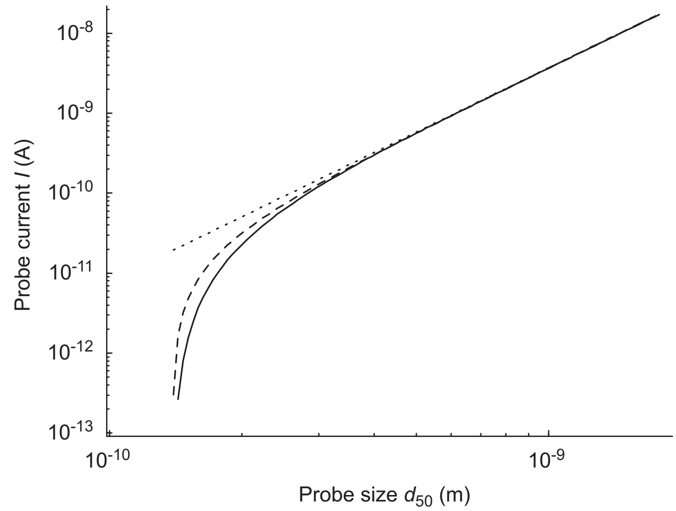

Fig. 1. Three curves relating the beam current I to d_{50}, with microscope parameters summarized in Table 1. The solid curve is derived from Eq. (1) with C_c=1 mm. The dotted curve depicts the approximation in Eq. (3), and the dashed curve shows the approximation given by Eq. (5) and used throughout the paper.

Table 1
Microscope parameters used in the simulations.

E_0	λ	C_s	B_r
300 kV	1.97 pm	1 mm	5×10^7 A m^{-2} sr^{-1} V^{-1}

Various terms in Eq. (1) can be neglected for certain microscope settings. This is shown, for instance, in Ref. [12], where various approximations for low values of E_0 are given. In this paper, a similar derivation is given. The acceleration voltage E_0 equals 300 keV, while δE is only 0.4 eV, as a consequence the d_c^2-term can be neglected. In addition, numerical calculations showed that the angle α minimizing d_{50} for a given I increases monotonically with d_{50}. This suggests that, for large I and large d_{50}, the d_A^4-term can be dropped from the model as well. The model then simplifies to

$$d_{50}^{1.3}=d_I^{1.3}+d_s^{1.3}. \tag{2}$$

It can be shown analytically that, in this case, d_{50} is minimized with respect to α if

$$I=0.25\pi^2B_rE_0C_s^{-2/3}d_{50}^{8/3}. \tag{3}$$

This function is shown as the dotted line in Fig. 1. It is clear that for large probe sizes, it provides an excellent approximation to the original model. For smaller probe sizes, however, the simplified model behaves qualitatively different from the exact model.

A better approximation is obtained by incorporating the geometrically limited probe size $d_{g,50}$. It is defined as the probe size in the limit of zero probe current. For higher values of E_0, it is approximated by

$$d_{g,50}^4=d_A^4+d_s^4.$$

Analytically minimizing $d_{g,50}$ with respect to α yields

$$d_{g,50}=0.47C_s^{1/4}\lambda^{3/4}. \tag{4}$$

By definition it holds that I tends to 0 if d_{50} approaches $d_{g,50}$. We therefore propose to incorporate $d_{g,50}$ in Eq. (3) in the following way:

$$I=0.25\pi^2B_rE_0C_s^{-2/3}(d_{50}^{8/3}-d_{g,50}^{8/3}). \tag{5}$$