Quantitative High-Resolution TEM/STEM and Diffraction

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Atomic number estimation from STEM images: what are the limits?

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Statistical parameter estimation theory is a quantitative method to measure unknown structure parameters from electron microscopy images. Such images can be considered as data planes from which structure parameters can be determined. A parametric statistical model of the observations is needed in order to perform computer simulations of the images. This model needs to describe the electron-sample interaction, microscope transfer and image detection. The unknown parameters are then estimated by fitting the model to the experimental images using a criterion of goodness of fit [1]. Since the precision with which the parameters can be estimated is limited by noise, the goal of experiment design is to investigate which microscope settings are expected to yield the highest precision with which the structure parameters can be estimated. Statistical parameter estimation theory can be used for this purpose. Previous work has shown the Cramér-Rao Lower Bound (CRLB) to be a very efficient tool to compute the optimal experiment design that provides maximum precision [2-4]. The CRLB is a lower bound on the variance of unbiased estimators of the unknown parameters and can be computed from the parameterized probability (density) function of the observations. However, it is only defined when the probability (density) function is continuously differentiable with respect to the parameters. An alternative approach using the principles of detection theory [5] is therefore investigated for problems concerning the estimation of so-called restricted (or, discrete) parameters.

This alternative approach is needed for instance when considering the problem of identifying the atomic number Z from a Scanning Transmission Electron Microscopy (STEM) image. Note that chemical theory restricts the atomic number to be a positive integer, which makes it is a restricted parameter. A priori knowledge about the atom types that may be present in a sample and their concentration ratios is usually available. In such cases, the question reduces to distinguishing between a finite plausible set of values for the atomic numbers, given the experimental STEM observations. Here we restrict to the problem of deciding between two hypotheses, where each hypothesis corresponds to the assumption of a specific Z value: H_0: $Z=Z_0$ and H_1: $Z=Z_1$. H_0 is referred to as the null hypothesis and H_1 as the alternative hypothesis. Following the so-called Bayesian approach, prior probabilities $P(H_0)$ and $P(H_1)$ are assumed known. In this way, we express a prior belief in the likelihood of the hypotheses. If the presence of an atom of type Z_0 or Z_1 is equally likely, then it is reasonable to assign equal probabilities of 1/2. An expression for the probability of error P_e can now be defined as the sum of the probabilities of deciding H_i if H_j is true, where the two possible errors are weighted appropriately to yield an overall error measure. For the computation of P_e a simplified discrete parametric model of an isolated atom is derived, assuming Poisson noise statistics. Decision rules are now defined such that P_e is minimized. For this purpose, it is shown in [5] that for equal prior probabilities, we should then decide H_1 if the conditional joint probability function given Z_1 evaluated at the available observations, is greater than that given Z_0. This corresponds to choosing the hypothesis for which the log likelihood function is maximal. The difference between the log likelihood function for Z_1 and this for Z_0 is defined as the log likelihood ratio. Using repetitive simulations, P_e can be computed for different experimental settings, in order to compute which experimental settings minimise the probability to assign an incorrect hypothesis.

A tightly connected performance measure that is investigated as a possible alternative to optimize the experiment design, is based on the Kullback-Leibler divergence [7,8]. This measure quantifies the difference between two probability distributions, circumventing the need for excessive image simulations. The sum of Kullback-Leibler divergences corresponds to the difference of the expected or mean log-likelihood ratio under H_1 and the corresponding value when assuming H_0 to be true.

An experimental case study is presented to show the practical use of these proposed measures for the optimisation of the experiment design. More precisely the optimal inner detector radius of an annular detector is derived when deciding between the presences of an Al or Ti atom in STEM

images, such as in the experimental study in [6]. Simulations are performed under both hypotheses H_0: $Z=Z_0=13$ (Al) and H_1: $Z=Z_1=22$ (Ti). For every simulation experiment the log likelihood ratio is calculated and so Figures 1-3 are obtained for three different inner detector radii (0.7 $Å^{-1}$, 1.1 $Å^{-1}$ and 2.5 $Å^{-1}$). It is found that the sum of Kullback-Leibler divergences and the probability of error provide consistent results. Indeed, the probability to assign the wrong hypothesis decreases when the distributions of the log-likelihood ratio under the considered hypotheses are better separated, and thus when the sum of Kullback-Leibler divergences increases. It is seen in Figure 4 that a minimum for the probability of error is found for the inner detector radius maximising the sum of Kullback-Leibler divergences, namely for 1.1 $Å^{-1}$. We see in Figures 1-3 that for this inner detector radius the log-likelihood functions under H_0 and H_1 are separated the most. This proves the Kullback-Leibler divergence to be an efficient alternative performance measure to optimize the experiment design.

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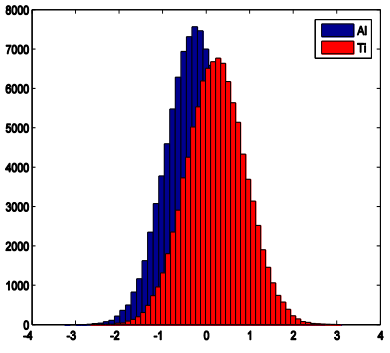

Figure 1. Log likelihood ratio for inner detector radius 0.7 $Å^{-1}$ for Al (blue) and Ti (red)

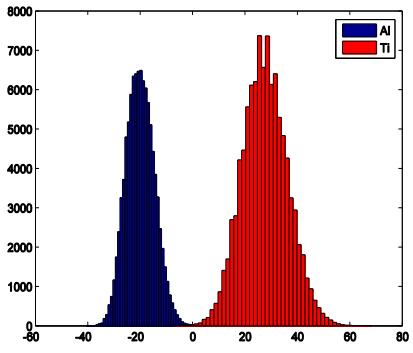

Figure 2. Log likelihood ratio for inner detector radius 1.1 $Å^{-1}$ for Al (blue) and Ti (red)

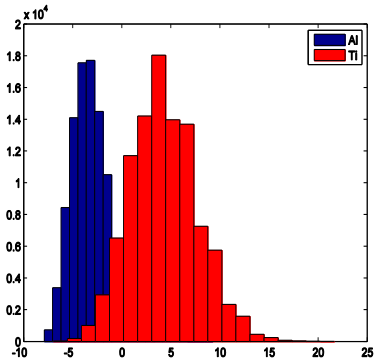

Figure 3. Log likelihood ratio for inner detector radius 2.5 $Å^{-1}$ for Al (blue) and Ti (red)

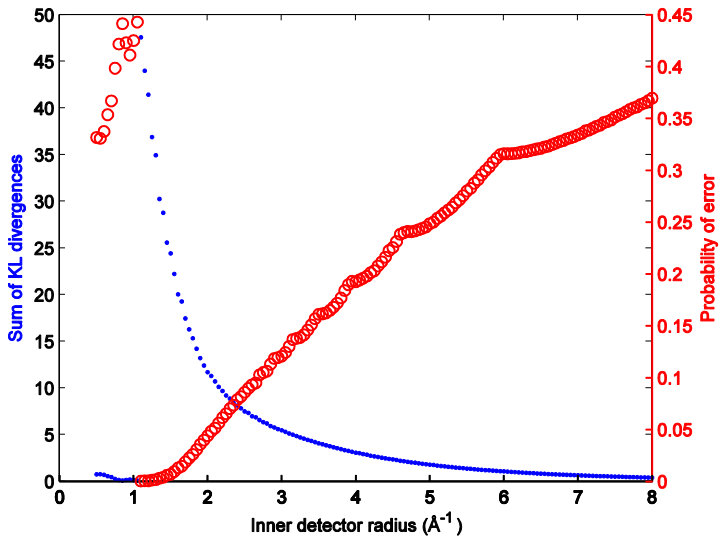

Figure 4. Probability of error (red) and sum of KL divergences (blue) for Al and Ti as a function of the inner detector radius ($Å^{-1}$).