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The Probe Profile and Lateral Resolution of Scanning Transmission Electron Microscopy of Thick Specimens

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Abstract

Lateral profiles of the electron probe of scanning transmission electron microscopy (STEM) were simulated at different vertical positions in a micrometers-thick carbon sample. The simulations were carried out using the Monte Carlo method in the CASINO software. A model was developed to fit the probe profiles. The model consisted of the sum of a Gaussian function describing the central peak of the profile, and two exponential decay functions describing the tail of the profile. Calculations were performed to investigate the fraction of unscattered electrons as function of the vertical position of the probe in the sample. Line scans were also simulated over gold nanoparticles at the bottom of a carbon film to calculate the achievable resolution as function of the sample thickness and the number of electrons. The resolution was shown to be noise limited for film thicknesses less than 1 μm . Probe broadening limited the resolution for thicker films. The validity of the simulation method was verified by comparing simulated data with experimental data. The simulation method can be used as quantitative method to predict STEM performance or to interpret STEM images of thick specimens.

Keywords

Electron probe broadening; Monte Carlo simulations; nanoparticles; noise-limited resolution; STEM; spatial resolution; thick specimen

Introduction

STEM can be used for nanometer-resolution imaging of nanoparticles and biological structures in micrometers-thick organic (Hyun, et al., 2008; Loos, et al., 2009; Miyazawa, et al., 2003; Sousa, et al., 2008) or liquid specimens (de Jonge, et al., 2009). For the imaging of micrometer-thick samples, the spatial resolution is not limited by the electron optics, as is the case for ultrathin amorphous substrates, but by noise or by beam broadening caused by elastic scattering of the electron beam in the specimen (de Jonge, et al., 2010; Reimer & Kohl, 2008). Analytical expressions were obtained for the lateral width, where the broadening on account of (multiple) electron scattering was regarded as a Gaussian distribution for thin films (Doig & Flewitt, 1982; Doig, et al., 1981; Gentsch, et al., 1974; Hall, et al., 1981; Reimer & Kohl, 2008). However, it is not clear if the Gaussian distribution for the lateral probe shape is still valid for samples much thicker than the mean-free-path-length for elastic scattering. Moreover, the electron probe may exhibit a more

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complex shape than described by a single Gaussian-like function. Furthermore, the variation of the probe shape as a function of the vertical position of the electron probe in thick sample was not studied in detail. For example, Boltzmann transport calculations (Groves, 1975; Rez, 1983), and Monte Carlo simulations were used to study the electron probe at the bottom surface of samples (Hyun, et al., 2008; Sousa, et al., 2009), but the evolution of probe profile in micrometer-thick sample was not carried out.

Here, we present a detailed study of the lateral electron probe profile using Monte Carlo simulations for specific sample geometries and STEM settings. Different analytical models will be discussed to describe the effect of beam broadening on the probe profile, including the effect that the unperturbed probe is still present until the probe has reached a certain depth in the sample. In addition, the achievable spatial resolution in thick specimens will be evaluated as function of the vertical position within the sample, and of the amount of electrons in the probe, to test noise-limited resolution. Finally, we will compare simulated data with data obtained from aberration corrected STEM of a test sample.

Methods

Simulation of Electron Position in a Thick Sample

The broadening of the electron probe inside a micrometers-thick film was studied by calculating the lateral electron positions at different vertical positions using Monte Carlo simulations with the program CASINO (Demers, et al., 2010). The software included the ELSEPA elastic electron scattering cross section (Salvat, et al., 2005), Poisson noise characteristics of the electron source, and the electron optics of STEM (annular dark field detector and scanning of a focused electron beam). The simulation of STEM images was calibrated using experimental data (de Jonge, et al., 2010; Demers, et al., 2010). In CASINO, a sample was defined by combining different geometric shapes. Each shape defined the boundary between two regions. The program generated a virtual collision each time an electron trajectory intercepted a region boundary. Horizontal planes were added to the sample at different vertical positions. Each time an electron crossed a plane, the electron position and the type of collision were recorded, such that a 3D model of the beam broadening was generated.

In this study, the beam broadening was simulated using a 5 μm -thick sample and 100,000 electron trajectories except when mentioned otherwise. The number of trajectories was chosen as a compromise between the statistical error in the results and the size of the resulting data file. A total of 51 horizontal planes was placed at different vertical positions z : from 0 nm to 5 μm by 100 nm steps. For the convergent probe, the focal position was changed to correspond to the vertical position for each data point, i.e., the electron profile was always calculated in focus position at the smallest probe size of a convergent probe. A Gaussian beam with a diameter of $d_G = 1$ nm was used. This diameter contained 99% of all electrons and was related to the standard deviation σ of the Gaussian function by $d_G = 4.576 \sigma$.

The lateral probe shape $N(r, z)$ was calculated from the simulated electron positions. A script was used to extract the electron position (x, y, z) at each vertical position (horizontal plane) for each electron trajectory. From the list of electron positions, a histogram of the radial distribution was calculated for each vertical position with a lateral step size of 0.1 nm. Because the areas associated with each radial position were not uniform, the number of electrons was transformed into the electron density for each radial position. For each radial position, the electron density was calculated by the number of electrons, divided by the area. The histogram of electron density was then used to determine the diameter of the probe. Firstly, the maximum value of the central peak was determined and then the lateral positions

x_1 and x_2 on either side of the peak with half maximum value were determined. The size d_{FWHM} was calculated from the difference of these values: $d_{FWHM} = |x_2 - x_1|$.

At each vertical position, the number of collisions (elastic scattering) undergone by an electron was also calculated. This number of elastic scatterings allowed separating unscattered from scattered electrons contributions to the probe profile.

Fit of the Lateral Probe Profile

A model was developed to describe the lateral electron probe profile. A profile was fitted using the least squares maximum likelihood estimator method using a modified Levenberg-Marquardt algorithm (Press, et al., 2002). To assure a good convergence, the lateral profile at a vertical position (z) of zero (top of the sample) was first fitted using the initial Gaussian probe parameters as input parameter. The electron probe did not suffer any broadening at this vertical position. The other profiles were fitted in order of increasing z and using the previous obtained fit parameters as input parameters. The residual, defined as the difference between the data and the model, plot was used to evaluate the agreement of the fitted model over the entire range of data.

Simulations of Gold Nanoparticle Line Scans

STEM line scans of gold nanoparticles at the bottom of a sample were simulated to evaluate the achievable spatial resolution. The varied conditions were the sample thickness T , the nanoparticle diameter d , and the number of electrons per pixel N . The nanoparticle diameters were varied until a signal-to-noise-ratio (SNR) of 5 was reached. A range of diameters of 0.2 to 300 nm was needed to cover the studied conditions, which implied that a rather large number of simulations was needed to find all gold nanoparticle diameters. We reduced the simulation time by using only two scan points to calculate the SNR for each diameter. The first scan point was directly over the nanoparticle and provided its maximum value N_M . The other scan point was 1 μm adjacent to the nanoparticle and corresponded to the background value N_B . From these values, the SNR was calculated as:

$$SNR = \frac{N_M - N_B}{\sqrt{N_M + N_B}}, \quad (1)$$

where we assumed that the noise exhibited a normal distribution (Rose, 1948a). The two simulated points were repeated 10 times for each diameter to obtain an average value of the SNR. The diameter corresponding to $SNR = 5$ was determined from a plot of SNR versus diameter.

After the diameter was determined, a line scan over the nanoparticle was simulated. The FWHM was calculated from this line scan. The half-maximum (HM) value was calculated by $HM = N_B + (N_M - N_B)/2$, and the two positions x_1 and x_2 on each side of the central peak were obtained. The FWHM was given by the difference of these positions $FWHM = x_2 - x_1$ (Demers, et al., 2010). The line scan simulation was repeated 10 times to obtain an average value of FWHM.

Sample Preparation

A test sample was prepared by dispersing gold nanoparticles of various sizes on either side of a standard silicon nitride (SiN) membrane (SPI) of thickness 500 nm. A solution containing gold nanoparticles of sizes 2, 5, 10, and 30 nm (Ted Pella) diluted in ethanol was mixed in an ultrasonic bath for 2 minutes. This solution was applied on either side of the SiN membrane. The sample was air-dried for 3 minutes, and then plasma cleaned for 30 seconds on both sides of the membrane.

Electron Microscopy

Electron microscopy was performed using an aberration corrected STEM (JEOL 2200 FS) operated at a beam energy of 200 KeV. The images were obtained at a probe convergence semi-angle of 41 mrad, a probe current of 83 pA, a magnification of 500,000, a pixel size of 0.56 nm, a pixel dwell time of 32 μ sec, and using the high angle annular dark field (HAADF) detector. It was observed that the beam broadening caused images obtained at the bottom of the membrane to be considerably blurred, making it difficult to locate its exact focus position. To overcome this limitation, a focal series dataset consisting of 100 images, each differing in focus by 8 nm was obtained, from which the best focus position was selected. For details on the sample and the STEM images see elsewhere (Ramachandra, et al., 2011).

Results and Discussion

Fit Lateral Profile

When a narrow electron beam travels through a material it broadens on account of elastic scattering of the electrons with the atoms of the sample, leading to changes of the directions of the electrons. As the electron beam propagates deeper in the sample, the initial probe profile in vacuum decreases in intensity, and a broad beam tail forms. The broadening of the electron probe was studied by calculating the lateral electron positions at different vertical positions using Monte Carlo simulations with software calibrated using experimental STEM data (de Jonge, et al., 2010; Demers, et al., 2010). Figure 1a shows the lateral profile of an electron beam at a vertical position of 1 μ m in a carbon sample of 5 μ m thickness. The incident electron energy was 200 keV. For ease of calculation a parallel beam was used (semi-angle of 0 mrad), which differs from STEM in reality but is an acceptable method to study beam broadening caused by scattering, since this type of beam broadening does not differ between a convergent and a parallel beam (see below).

The profile was fitted to a general model based on a Gaussian function developed by Doig and Flewitt (Doig & Flewitt, 1982; Doig, et al., 1981). In this model, the electron intensity distribution $N_G(r, z)$ at a distance r and vertical position z in the film was approximated by:

$$N_G(r, z) = N_0 \frac{1}{\pi \cdot (2\sigma^2 + \beta z^3)} \exp \left[-\frac{r^2}{2\sigma^2 + \beta z^3} \right], \quad (2)$$

where N_0 is the number of incident electrons, σ is a measure of the incident electron probe size with probe diameter $d_{FWHM} = 2.35\sigma$, and β is a parameter defining the electron scattering characteristics of the film material given (in units of nm^{-1}) by:

$$\beta = \left(\frac{4Z}{E_0} \right)^2 \cdot \frac{\rho}{A} \cdot 500, \quad (3)$$

here, Z , A , and ρ are the atomic number, the atomic weight, and mass density of the film material, respectively, and E_0 is the electron accelerating voltage in eV. As observed in Figure 1a, the fit of this equation was not able to model the lateral profile. The residual plot shows that neither the central peak nor the large tail was correctly modeled.

Models of the probe broadening (Doig & Flewitt, 1982; Doig, et al., 1981; Hall, et al., 1981; Michael & Williams, 1987; Reed, 1982; Reimer & Kohl, 2008) often assume that the broadening can be described by the convolution of the Gaussian incident beam with another Gaussian to give a single Gaussian with the width given by the quadrature of the two Gaussian widths. The underlying thought of this hypothesis is that all electron trajectories

are broadened such that the initial beam (unscattered electron) completely disappears. As observed in Figure 1a, this is not the case. For this reason we have tested to fit a double Gaussian function N_{GG} to the simulated lateral profile:

$$N_{GG}(r, z) = A_1 \exp \left[-\frac{r^2}{2\sigma_1^2} \right] + A_2 \exp \left[-\frac{r^2}{2\sigma_2^2} \right], \quad (4)$$

where A is the area, and σ is the width of the Gaussian function. The first term (A_1) was used to model the Gaussian incident beam, and the second term (A_2) was used to model the broadened tail as observed in Figure 1b. The residual shows an improvement when using the N_{GG} model at the center of the lateral profile. But large differences were still present in the tail away from the center.

Most models of probe broadening furthermore assume that the broadening can be described by a Gaussian function. A different function is apparently needed to model the broadened tail of the profile. Boltzmann transport calculations (Rez, 1983) and Monte Carlo simulations (Williams, et al., 1992) shown that the non-Gaussian probe broadening was caused by plural scattering effects. The Gaussian shape of the electron profile was due to the form of the Rutherford electron scattering model used in earlier calculations and not by the electron diffusion (Rez, 1983). Mott electron scattering models (Czyzewski, et al., 1990; Jablonski, et al., 2003; Mott & Massey, 1965), like the one used in this work (Salvat, et al., 2005), could lead to non-Gaussian probe broadening (Rez, 1983). We have found empirically that a model fitting the simulated data is composed of three terms:

$$N_{GEE}(r, z) = A_G \exp \left[-\frac{r^2}{2\sigma^2} \right] + H_1 \exp \left[-\frac{|r|}{\tau_1} \right] + H_2 \exp \left[-\frac{|r|}{\tau_2} \right], \quad (5)$$

where A_G is the area, σ is the width of a Gaussian function, and two exponential decay functions are composed of the height factor H , and decay period τ . Figure 1c shows a fit of this model to the simulated lateral probe at a vertical position of 1 μm . The obtained residuals were distributed randomly around zero and did not show any pattern, which indicated that the fitted model described the data correctly. Other functions were also tested, but they did not correctly fit the central peak and tail at the same time. These functions included: the addition or product of two Gaussian functions, a convolution of a Gaussian and Lorentzian, and a Pseudo-Voigt function.

The function can be understood as follows. The Gaussian describes the original unperturbed probe. The initial probe exhibited a Gaussian distribution with a diameter d_G of 1 nm ($\sigma = d_G/4.576 = 0.22$ nm). A Gaussian function was used to model the central peak. Even at z of 1 μm the central peak fitted a Gaussian function with the same width as the original probe profile. It was found that this central peak mainly contained unscattered electrons. The fit of the Gaussian function was repeated on the lateral profile composed of the unscattered electrons only. Similar fitting parameters of the Gaussian function were obtained. Figure 1d shows that the magnitude of this peak decreased with the vertical position until it completely disappeared after a critical depth in the sample, since the probability for an electron to experience one or more elastic collisions increases rapidly with the vertical position of the electron.

The first exponential is needed to obtain a transition between the central peak and the large tail (a Gaussian and a single exponential did not fit the data correctly). The second exponential decay function describes the extended large tail of the profile and become more important with increasing vertical position. We propose that the two exponential functions

represent the transition between single and multiple-scattering regime of the electron trajectories.

Probe Size in a Thick Sample

We have studied the probe size for different vertical positions. Figure 2 shows that the probe size defined as the FWHM of the probe profile d_{FWHM} increases with the increase of vertical position. The size d_{FWHM} was also calculated for a convergent probe of 41 mrad. In that case, the focal position z_f as used in CASINO was changed for each data point and set equal to the vertical position z . The d_{FWHM} was then calculated for each combination of focal point and vertical position ($z_f = z$). As shown in Figure 2, no difference in the probe broadening effect was observed between a parallel probe and a convergent probe. Thus, the broadening effect does not depend on the incident probe semi-angle at the focus position but the vertical position in the sample does affect the probe size.

A comparison of the lateral probe size obtained with a probe broadening model proposed by Reimer (Reimer & Kohl, 2008) is also shown in Figure 2. This probe-broadening model, which assumes a Gaussian probe profile for the broadening, has a dependency on the vertical position (thickness T in nm), energy E and sample composition. With this model, the d_{FWHM} in nm is given by

$$d_{FWHM} = 2\sqrt{\ln 2} \cdot 1.05 \times 10^{12} \sqrt{\frac{\rho}{A} \frac{Z}{E} \frac{1+E/E_0}{1+E/2E_0}} T^{3/2}, \quad (6)$$

where the factor $2\sqrt{\ln 2}$ is used to transform the Gaussian sigma factor of the original equation in Reimer (Reimer & Kohl, 2008) into the FWHM of the Gaussian, ρ is the mass density, A is the atomic weight, Z is the atomic number of the sample, E is the electron energy in keV, E_0 is rest energy, and T in nm is the thickness of the sample. Apparently, the Reimer model over-estimates the probe size at lower thicknesses and under-estimates the probe size at larger thicknesses.

The role of the sample composition in the probe broadening was studied by repeating the simulation for a gold sample. Figure 2 shows that the evolution of d_{FWHM} is similar for both gold and carbon samples, but the probe size obtained for gold is a factor of ten larger than for carbon. This factor was obtained by averaging the ratio of $d_{FWHM, Au}/d_{FWHM, C}$ for all vertical positions. To understand this factor, the ratio of both mean-free-path-lengths was calculated. The mean-free-path-length λ represent the average length an electrons travel before it undergoes an elastic collision, and is calculated from the total elastic cross section σ_{el} as (Reimer & Kohl, 2008):

$$\lambda = \frac{A}{N_A \cdot \rho \cdot \sigma_{el}}, \quad (7)$$

with Avogadro's number N_A . With the ELSEPA model (Salvat, et al., 2005) implemented in the CASINO software, values of 152 and 12 nm, were obtained at 200 keV, for carbon and gold, respectively, and a ratio $\lambda_C/\lambda_{Au} = 13$ was calculated. Note that a slightly larger ratio of 14 was obtained using the Rutherford cross section (Reimer & Kohl, 2008) instead of the ELSEPA model. These ratios were of the same order as the ratio of ten obtained between gold and carbon in Figure 2.

Fraction of unscattered Electrons

For a sample thinner than several λ s, the probe size d_{FWHM} may not be the correct estimate of the lateral resolution as shown previously (de Jonge, et al., 2010; Reimer & Kohl, 2008). The central peak of the probe profile composed of the unscattered electrons still provides the highest resolution when imaging a specimen, while the broadened beam tails contribute to an increased noise level with respect to a very thin sample. How the Gaussian probe profile is preserved with the vertical position was studied using the lateral probe profile of the unscattered electrons. The simulation software allowed us to extract the lateral profile of the unscattered electrons for different vertical positions. These unscattered electrons preserve the initial Gaussian profile and size, but the number of electrons in the profile decreases with increasing depth in the sample, as shown in Figure 3a for both a gold and a carbon sample. A more rapid decrease was observed for the gold sample on account of the smaller λ of a gold atom versus a carbon atom. Figure 3b shows the fraction of unscattered electrons as function of the vertical position normalized to λ . It can be seen that the fraction of unscattered electrons (fraction of the initial probe) was independent of the sample composition with this normalization. A universal model for the fraction of unscattered electrons was developed from the simulated data. The fraction of unscattered electrons data from H_2O , C, SiN, and Au samples were fitted with an exponential decay function. The fraction of unscattered electrons f_e is given by

$$f_e = 96 \cdot \exp\left(-\frac{z/\lambda}{1.08}\right). \quad (8)$$

This model can be used to find the fraction of electrons that will contribute to the imaging signal without loss of lateral resolution.

Effect of the Noise

The spatial resolution is not only limited by the probe size, but also by the statistical noise related to the number of electrons used for imaging. To study the influence of the noise on the resolution in combination with the effects of beam broadening we have simulated lateral line scans over gold nanoparticles at the bottom of a carbon sample. The resolution was evaluated on the basis of the Rose criterion stating that a pixel is detectable in a noisy background if the signal-to-noise-ratio (SNR) is at least 5 (Rose, 1948b). A series of simulations was performed (see Table 1) varying the specimen thicknesses, and number of electrons in the probe. For each condition, a line scan was simulated with a nanoparticle diameter d optimized to obtain $SNR \cong 5$, from which a FWHM was calculated, leading to the value $d_{N, FWHM}$, with N the number of incident electrons.

Figure 4 shows examples of the interdependence between the nanoparticle diameter, the sample thickness, and the number of electrons. In Figure 4a and b, a nanoparticle with a diameter of 5 nm is compared for two thicknesses: 0.2 and 0.5 μm . The number of electrons was increased by a factor 10 for the thicker sample to satisfy $SNR = 5$, leading to a factor of 25 larger background signal from the carbon film. Interestingly, the FWHM did not increase significantly. The increase of the background was caused by both a factor of ten more electrons, and an increasing number of scattered electrons in the thicker substrate leading to additional signals in the ADF detector. If the number of electrons is to be kept constant, the nanoparticle diameter can be increased for thicker sample to satisfy $SNR \cong 5$, as shown in Figure 4c and d. A larger diameter was needed for the thicker film to satisfy the Rose criterion.

Figure 5 shows the probe size $d_{N, FWHM}$ in a carbon film obtained over gold nanoparticles of such diameters that the condition $SNR \cong 5$ was satisfied. These simulated data points $d_{N, FWHM}$ represent the achievable spatial resolution as function of the sample thickness and the amount of electrons in the probe, accounting for the effects of beam broadening and statistical noise. The spatial resolution is always smaller (larger in number) than the probe size d_{FWHM} . For a maximal number of electrons the resolution approaches the curve of d_{FWHM} , which is the limit due to probe broadening only. It is not possible to improve the resolution beyond the limit of the probe size by increasing the electron dose. The spatial resolution is purely noise-limited for the thinner samples imaged with the lowest number of electrons. The highest resolution can be achieved for thin samples ($<0.2 \mu m$), where the probe profile still contains a significant portion of unscattered electrons with a narrow shape. The resolution obtained on nanoparticles positioned below a micrometers-thick sample is limited by beam broadening. The resolution-limiting factor transitions in the intermediate regime ($0.2 - \sim 1 \mu m$).

Note that nanometer resolution can be achieved for the imaging of nanoparticles on top of the material even for micrometers-thick specimens and that the resolution is noise limited in this case (de Jonge, et al., 2010). The nanoparticle line scan results presented in Figure 5 were obtained for a parallel beam. It was confirmed by simulations that no difference in $d_{N, FWHM}$ values were obtained with a convergent beam semi-angle of 41 mrad. Like the probe broadening (see Figure 2), the nanoparticle lateral resolution does not depend on the converge angle at the focal plane.

An important question is what happens for a nanoparticle at a certain vertical position within the specimen not at the bottom or the top. The probe shape at the corresponding vertical position is given by d_{FWHM} , which determined the maximum achievable resolution at that depth. Once a nanoparticle is scanned by the STEM probe the scattered electrons have to travel through the remaining material below the nanoparticle to the ADF detector, which introduces additional scattering events. These events do not decrease the maximal achievable resolution, but rather decrease the SNR and thereby the actual resolution $d_{N, FWHM}$. In essence, imaging a nanoparticle with material below it is equivalent to imaging with a lower N . Figure 5b shows the dependence of $d_{N, FWHM}$ on the vertical position for different sample thicknesses T . The nanoparticle diameters were chosen to achieve $SNR \cong 5$. The thickness of the material below the nanoparticle degraded the resolution only in the noise-limited regime. Probe broadening presented the sole limitation for the thicker samples.

Comparison with experimental data

In order to verify that the simulated data corresponds to real STEM images, we have compared experimental STEM data obtained on gold nanoparticles at the bottom surface of a SiN film of 500 nm thickness with simulated data in Figure 6a. A ten-pixel-wide line scan was extracted from a STEM image at the location of a gold nanoparticle, and the data was averaged over the line width to reduce the noise (corresponding to 1.7×10^5 electrons per data point). To be able to compare the shapes of the line scans, the line scans were scaled to zero at the lowest value and normalized to unity at maximum intensity. The comparison of both line scans demonstrates that the Monte Carlo simulations correspond to the experimental data. The simulation method presented here is thus a valid quantitative method to predict the influence of beam broadening on the shape of line scans of nanoparticles at bottom surface of a thin film.

The probe shape model N_{GEE} was fitted to the simulated line scan as shown in Figure 6b. For a line scan over a nanoparticle, the shape was not only described by the probe shape, but also by the geometry of the nanoparticle. The Gaussian function of the model was used to fit

the central peak, representing the shape of the nanoparticle, and one exponential decay function was needed to model the tails on each side of the peak. The first exponential was found to be zero in this fit. The scattering/multiple-scattering in the gold (around 90% of the electrons are scattered in the nanoparticle) should be the same for the tail and the center of the probe. Imagine the shape in Figure 1 scanning across the particle; at the center of the nanoparticle the same fraction of the electrons will be scattered into the ADF detector. But because the nanoparticle diameter is larger than the central portion of the probe shape, we only see the part of the multi-scattering exponential decay function (H_2 and τ_2 from eq. (5)) larger than the diameter in the line scan.

This result shows that even after the convolution of the probe shape with the nanoparticle and the substrate, the exponential function observed in the probe shape was still present. The N_{GEE} model derived in this study thus seems to represent a general model to describe the lateral shape of a focused STEM electron beam in a sample of such thickness that beam broadening occurs.

Conclusions

We have used Monte Carlo simulations to study the lateral probe profile of STEM in micrometers-thick samples. It was found that the probe profile was best described by a Gaussian and two exponential decay functions describing extended beam tails. The lateral probe size depends on the vertical position and the sample composition, but not on the incident probe semi-angle at the focus position. The fraction of unscattered electrons is independent of the sample composition after normalization of the vertical position by the mean-free-path length. Simulations of line scans over gold nanoparticle positioned at the bottom of a carbon film have shown that the lateral resolution is noise limited for film thicknesses $< 0.2 \mu\text{m}$, and by the probe broadening for film thicknesses $> 1 \mu\text{m}$; the intermediate thicknesses represent a transition regime. The simulation method is calibrated for real STEM imaging, as was demonstrated for the imaging of a gold nanoparticle below a 500 nm-thick SiN membrane.

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Figure 1.

Lateral profile $N(r,z)$ of the electron probe versus the distance from the central beam axis r at a vertical position $z = 1 \mu\text{m}$ within the specimen. Profile obtained for a parallel electron beam at 200 keV in a $5 \mu\text{m}$ -thick carbon sample. Blue dots are simulated data, and green line is the model fitted to the data. (a) Simulated probe fitted with a single Gaussian model N_G as derived by Doig and Flewitt (Doig & Flewitt, 1982; Doig, et al., 1981). (b) A double Gaussian function N_{GG} was used to model the probe. (c) A Gaussian function (G) was used to model the unscattered probe (central peak), and two exponential decay functions (E_1 and E_2) were used to model the probe broadening (scattered tail) N_{GEE} . The relative residual R of the fit is shown at the bottom of each graphic. (d) Variation of the magnitude of the components of the fitted N_{GEE} model with the vertical position.

Figure 2.

Variation of the probe size as measured by the d_{FWHM} versus the vertical position. A 5 μm -thick carbon sample (green) was used. The probe size obtained from simulations for a parallel beam (green line), and a convergent probe (convergence semi-angle of 41 mrad, green triangles) are compared. For the convergent probe, the focal position was changed to correspond to the vertical position for each data point. The probe size for a simulated parallel probe for a gold sample (blue) is also included. Dashed lines are values of d_{FWHM} given by the Reimer model (Reimer & Kohl, 2008).

Figure 3.

Fraction of unscattered electrons for different vertical positions. Two sample compositions are used: a gold (blue) and a carbon (green). (a) Fraction of unscattered electrons versus the vertical position. (b) Fraction of unscattered electrons versus the vertical position normalized by the mean-free-path-length in each material ($\lambda_{\text{Au}} = 12 \text{ nm}$ and $\lambda_{\text{C}} = 152 \text{ nm}$).

Figure 4.

Simulated scanning transmission electron microscopy (STEM) line scans of gold nanoparticles at the bottom surface of a carbon film for a parallel beam. The nanoparticle diameters were chosen to obtain a signal-to-noise ratio (SNR) $\cong 5$ at an incident energy of 200 keV. (a,b) Nanoparticle diameter of 5 nm: (a) 1,000 electrons and a sample thickness of 0.2 μm and (b) 10,000 electrons and a sample thickness of 0.5 μm . (c,d) 10,000 electrons: (c) nanoparticle diameter of 1.8 nm and a sample thickness of 0.2 μm and (d) nanoparticle diameter of 31 nm and a sample thickness of 2 μm .

Figure 5.

The smallest observable ($SNR \cong 5$) gold nanoparticle $d_{N,FWHM}$ (symbols) below and inside a carbon sample of various thicknesses. The values of $d_{N,FWHM}$ were obtained from simulated STEM line scans over gold nanoparticles for different amounts of incident electrons N at an incident energy of 200 keV for a parallel beam. A comparison with the broadened probe size d_{FWHM} (blue line) as function of the vertical location in a 5 μm-thick sample is also included. (a) Nanoparticle below the sample for different amounts of incident electrons N . See also Table 1. (b) Nanoparticle inside the sample for different vertical positions z and sample thicknesses T with 10,000 electrons per pixel. See also Table 2.

Figure 6.

Experimental and simulated STEM line scans of gold nanoparticles at the bottom surface of a 500 nm-thick silicon nitride film. (a) Comparison of experimental (blue line) and simulated (green line) STEM line scans at incident energy of 200 keV with a convergence semi-angle of 41 mrad. A selected region of the original image is shown in the background. The line scan was obtained from the average of a 10-pixel wide line over the nanoparticles as indicated by the white rectangle. The number of electrons N for each line scans was scaled (minimum value equal zero) and normalized to one. A nanoparticle diameter of 34 nm was used for the simulation. (b) Fit of the simulated data with the N_{GEE} model (blue dots are simulated data points). The relative residual R of fit is shown at the bottom.

Table 1

Analysis of simulated STEM signal peaks in line scans over gold nanoparticles of diameter d positioned at the bottom surface (with respect to the electron probe propagation direction) of a carbon film of thickness T for different number of electrons per pixel N . The signal-to-noise-ratio SNR and the full-width-half-maximum FWHM are given for four different numbers of electrons per pixel N .

N	T (μm)	d (nm)	SNR	FWHM (nm)
10^3	0.2	10	6.2	7.8
	0.5	19	6.3	13
	1.0	43	6.7	38
	2.0	105	6.3	124
10^4	0.2	3.6	6.2	2.8
	0.5	9.8	6.1	9.2
	1.0	25	6.8	28
	2.0	63	6.4	108
5×10^4	0.2	2.1	6.2	1.6
	0.5	6.6	6.4	6.6
	1.0	18	6.7	24
	2.0	47	6.6	106
10^5	0.2	1.6	5.6	1.2
	0.5	5.3	5.9	6.0
	1.0	15	6.4	23
	2.0	40	6.2	110

Table 2

Analysis of simulated STEM signal peaks in line scans over gold nanoparticles of diameter d positioned at a vertical position z inside a carbon film of thickness T .

T (μm)	z (μm)	d (nm)	SNR	FWHM (nm)
0.5	0.2	5.0	6.6	4.0
	0.5	9.8	6.1	9.2
1.0	0.2	6.3	6.9	4.8
	0.5	11	6.3	9.5
	1.0	25	6.8	28
2.0	0.2	8.8	6.3	7.2
	0.5	15	6.5	11
	1.0	28	6.5	28
	2.0	63	6.4	108
5.0	0.2	33	6.6	24
	0.5	41	6.8	31
	1.0	55	6.3	44
	2.0	102	6.4	113