Addendum. Laplacian image contrast in mirror electron microscopy

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We extend the theory of Laplacian image contrast in mirror electron microscopy (MEM) to the case where the sample is illuminated by a parallel, collimated beam. This popular imaging geometry corresponds to a modern low energy electron microscope equipped with a magnetic objective lens. We show that within the constraints of the relevant approximations; the results for parallel illumination differ only negligibly from diverging MEM specimen illumination conditions.

1. Introduction

A recent paper by Kennedy et al. (2010) describes how, under certain conditions, images formed in mirror electron microscopy (MEM) can be interpreted in terms of the Laplacian of small height or potential variations across a sample surface. The specific MEM imaging geometry considered conforms to an experimental configuration where the incident electron beam is slightly divergent as a result of the anode aperture. This arrangement has been considered by several authors (Dyukov et al. 1991; Nepijko & Sedov 1997; Nepijko et al. 2001a, b, 2003, 2007; Nepijko & Schönhense 2010). It is a subset of non-parallel MEM specimen illumination considered in the literature (Barnett & Nixon 1967a; Luk’yanov et al. 1974; Someya & Kobayashi 1974; Dupuy et al. 1984; Godehardt 1995). Modern low energy electron microscopes (LEEMs) are, however, equipped with a magnetic objective lens and it is customary to slightly converge the incident illumination to compensate for the diverging effect of the anode aperture, resulting in collimated illumination of the sample (Altman 2010; Tromp et al. 2010), within the domain of validity of the aperture lens approximation. This paper therefore considers how MEM Laplacian imaging theory (Kennedy et al. 2010) is modified by a parallel or collimated illumination geometry. We show, that in the limit of small objective lens defocus, the results converge to the original divergent illumination geometry considered by Kennedy et al. (2010), so that the conclusions drawn there remain valid.

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2. Comparison of divergent and parallel mirror electron microscopy illumination geometries

An electrostatic MEM immersion lens is shown schematically in figure 1, which illustrates the divergent MEM illumination geometry considered by Kennedy et al. (2010), Luk’yanov et al. (1974), Dyukov et al. (1991), Godehardt (1995), Nepijko & Sedov (1997), Nepijko et al. (2001a, b, 2003, 2007) and Nepijko & Schönhense (2010). Here, an electron beam of energy $U$ travels parallel to the optical axis $z$ of the immersion lens and passes through the grounded anode aperture $A$. The overall effect of the electric field in the vicinity of the grounded aperture is approximated by replacing the aperture with a thin diverging lens (Grant & Phillips 1990; Lenc & Müllerová 1992; Rempfer & Griffith 1992; Nepijko & Sedov 1997; Kennedy et al. 2010), so that the electron beam is deflected away from the optical axis $z$. The electron beam is therefore diverging as it interacts with the electric field above the specimen surface $C$, which is located a distance of $L$ from the anode and acts as the cathode of the immersion objective lens (Barnett & Nixon 1967b; Luk’yanov et al. 1974; Bok 1978; Bauer 1985, 1998; Kennedy et al. 2010). The cathode $C$ is held at a negative potential $V < -U/e < 0$ relative to the grounded anode, where $-e$ is the electronic charge, so that the electron beam is reflected in the vicinity of $z = L_M$, a distance of $\delta$ above the specimen surface,

$$L_M = L - \delta = -\frac{LU}{eV}, \quad (2.1)$$

and the returning electron beam is further deflected as it exits the anode aperture. For a perfectly flat, equipotential specimen, an electron that enters the anode aperture at $(x_0, y_0)$ is closest to the cathode surface at $(3x_0/2, 3y_0/2)$.

To a good approximation, the magnetic imaging part of the objective lens can be considered separately to the electrostatic MEM immersion lens of figure 1 (Bauer 1985, 1994). A virtual image is formed by retracing the exiting electron trajectories at $z = 0$ to the plane at $z = \Delta f + 4L_M/3$, which is the object plane of the magnetic objective lens defocused by $\Delta f$ (Rempfer & Griffith 1992; Nepijko & Sedov 1997; Kennedy et al. 2010). The magnification on this plane is

$$\bar{M}(\Delta f) = \frac{2}{3} \left( 1 - \frac{3\Delta f}{4L_M} \right), \quad (2.2)$$

compared with the specimen surface, which reduces to $2/3$ for small defocus.

In a modern LEEM instrument, it is customary to illuminate the specimen with an electron beam that is parallel to the optical axis $z$ (Altman 2010; Tromp et al. 2010). When treating the anode aperture $A$ as a diverging lens with focal length $-4L_M$ (Grant & Phillips 1990; Lenc & Müllerová 1992; Rempfer & Griffith 1992; Nepijko & Sedov 1997; Kennedy et al. 2010), we achieve parallel illumination by using a converging electron beam that is focused on the $z$-axis to the point $z = 4L_M$. After passing through the anode aperture the beam emerges parallel to the optical axis $z$. As shown in figure 2, for a perfectly flat, equipotential specimen an electron that enters the anode aperture at $(x_0, y_0)$ remains at $(x_0, y_0)$ in the vicinity of the turning region $z = L_M$, and exits the anode aperture at.
Figure 1. Classical electron trajectories (solid lines), travelling parallel to the optical axis $z$ in the electric field free region ($z < 0$) along the centre of an anode aperture $A$. The trajectories are deflected away from the $z$-axis both upon entering and exiting the anode–cathode region, owing to the distortion of the electric field in the vicinity of the anode aperture $A$. This effect is approximated by treating the aperture $A$ as a diverging lens (Grant & Phillips 1990; Lenc & Müllerová 1992; Rempfer & Griffith 1992; Nepijko & Sedov 1997; Kennedy et al. 2010). This deflection results in diverging illumination of the cathode specimen $C$, which is held at the potential $V < 0$ when compared with the anode, so that an electron of energy $U < -eV$ turns at a distance of $z = L_M$. The $y$-axis extends out of the page. Based on Kennedy et al. (2011). (Online version in colour.)

approximately the same transverse position. As before, we trace back along the apparent straight line path of the exiting electron to the virtual image plane $z = \Delta f + 4L_M/3$, where the virtual image plane is magnified by

$$M(\Delta f) = \frac{2}{3} \left(1 - \frac{3\Delta f}{8L_M}\right),$$

compared with the specimen surface.

Given the wide applicability of systems using the modern LEEM parallel illumination of the specimen (figure 2), it is important to consider the modifications to Laplacian imaging theory, since the latter was originally developed in a divergent illumination geometry.

3. Geometrical theory of mirror electron microscopy contrast

Following the approach of Kennedy et al. (2010), we consider the interaction of an electron with variations in electrical potential $V(x, y, \bar{z})$ above the specimen surface, where $\bar{z} = L - z$. This additional potential can be caused by areas of differing work function or applied voltage, and/or when a surface varies in height. Here, we concentrate on surface height variations, but the methodology is equally applicable to surface potential variations, or both height and potential variations. We approximate a surface with height variations, characterized by $H(x, y),$
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Figure 2. Classical electron trajectories (solid lines), in which the converging incident electron beam in the electric field free region \((z < 0)\) is focused on the optical axis \(z = 4L_M\). The trajectories are deflected away from the \(z\)-axis owing to the distortion of the electric field in the vicinity of the anode aperture \(A\). This effect is approximated by treating the aperture \(A\) as a diverging lens (Grant & Phillips 1990; Lenc & Müllerová 1992; Remper & Griffith 1992; Nepijko & Sedov 1997; Kennedy et al. 2010). The deflection of the converging beam away from the \(z\)-axis results in parallel illumination of the cathode specimen \(C\), which is held at the potential \(V < 0\) when compared with the anode, so that an electron of energy \(U < -eV\) turns at a distance of \(z = L_M\). The returning electron trajectories are deflected away from the \(z\)-axis when passing back out the anode aperture, retracing the incident trajectory for a perfectly flat equipotential specimen. As in figure 1, we trace back along the apparent straight line paths (dashed lines) to the virtual image plane in the vicinity of \(z = 4L_M/3\). The \(y\)-axis extends out of the page. Based on Kennedy et al. (2011). (Online version in colour.)

with the equivalent planar surface with a corresponding potential distribution (Nepijko & Sedov 1997)

\[
V(x, y, \bar{z} = 0) = \frac{VH(x, y)}{L}. \tag{3.1}
\]

Solving the Dirichlet problem for Laplace’s equation for a half space we have (Polozhiy 1967; Boudjelkha & Diaz 1972; Nepijko & Sedov 1997; Kennedy et al. 2010)

\[
V(x, y, \bar{z}) = \frac{\bar{z}}{2\pi} \int_{-\infty}^{\infty} \frac{V(\xi, \eta, \bar{z} = 0)}{((x - \xi)^2 + (y - \eta)^2 + \bar{z}^2)^{3/2}} \, d\xi \, d\eta, \tag{3.2}
\]

which expressed as a convolution is (Cowley 1995; Press et al. 2007)

\[
V(x, y, \bar{z}) = \frac{\bar{z}}{2\pi} V(x, y, \bar{z} = 0) \otimes (x^2 + y^2 + \bar{z}^2)^{-3/2}. \tag{3.3}
\]

From equation (3.1), the variation in electric potential above the specimen surface can then be expressed as the height function \(H(x, y)\) convolved with a smoothing function,

\[
V(x, y, \bar{z}) = \frac{\bar{z}}{2\pi L} H(x, y) \otimes (x^2 + y^2 + \bar{z}^2)^{-3/2}. \tag{3.4}
\]

As discussed in Kennedy et al. (2010), the smoothing function represents the blurring and softening of the electric field when moving away from the cathode surface. For example, the smoothed potential extends beyond the \((x, y)\) range of...
a localized hill or valley described by \( H(x, y) \). The geometrical theory of MEM contrast assumes that any change to the electron motion caused by the finite height variation of the specimen occurs very close to the sample surface, since the additional potential \( V(x, y, z) \) rapidly approaches zero as \( z \) increases away from the surface. Additionally, it is assumed that the \( z \)-dimensional motion is unchanged, so that all of the momentum change in the transverse dimensions \( (x, y) \) occurs very close to the classical turning point at \( z = L_M \).

In the modern LEEM geometry, an electron that enters the anode at \((x_0, y_0)\) is affected most strongly by the cathode at \((x_0, y_0)\) where it is closest to the surface (figure 2), rather than \((3x_0/2, 3y_0/2)\) for the divergent illumination geometry. We therefore estimate the \( x \) and \( y \) derivatives of the potential by integrating along the \( z \)-axis for the column \((x_0, y_0)\), obtaining the change to the \( x \) and \( y \) velocities, respectively. The shift of electron position \( S_x, S_y \) on the plane \( z = 4L_M/3 + \Delta f \) owing to \( H(x, y) \), which is scaled by \( 1/M \) (equation (2.3)) (Dyukov et al. 1991; Nepijko & Sedov 1997), is given by

\[
S_x(x, y, \delta, \Delta f) = \left( \frac{\partial}{\partial x} \right) \sqrt{\frac{L_M}{\pi}} \frac{9\Delta f}{8L_M - 3\Delta f} H(x, y) \otimes ((\delta^2 + x^2 + y^2)^{-3/4} \times (2E_E(x, y, \delta) - E_K(x, y, \delta)))
\]

and

\[
S_y(x, y, \delta, \Delta f) = \left( \frac{\partial}{\partial y} \right) \sqrt{\frac{L_M}{\pi}} \frac{9\Delta f}{8L_M - 3\Delta f} H(x, y) \otimes ((\delta^2 + x^2 + y^2)^{-3/4} \times (2E_E(x, y, \delta) - E_K(x, y, \delta)))
\]

where

\[
E_E(x, y, \delta) = E \left( \frac{1}{2} - \frac{\delta}{2(\delta^2 + x^2 + y^2)^{1/2}} \right)
\]

and

\[
E_K(x, y, \delta) = K \left( \frac{1}{2} - \frac{\delta}{2(\delta^2 + x^2 + y^2)^{1/2}} \right)
\]

and \( K, E \), respectively, denote complete elliptic integrals of the first and second kind (Abramowitz & Stegun 1964; Borwein & Borwein 1987). For \( \Delta f = 0 \), the electron shifts are zero, even for a rough surface with non-zero \( H(x, y) \). Therefore, the plane \( z = 4L_M/3 \) corresponds to the in-focus plane of minimum contrast, and a finite defocus \( \Delta f \) is required to obtain image contrast (Kennedy et al. 2010).

We note that equations (3.5) and (3.6) differ from equations (1.5) and (1.6) of Kennedy et al. (2010) only in that the term \( 9\Delta f/(8L_M - 6\Delta f) \) has been replaced by \( 9\Delta f/(8L_M - 3\Delta f) \) here. This is equivalent to multiplying the position shift functions in the divergent illumination geometry by the change in magnification \( \tilde{M}/M \) (equations (2.2) and (2.3), respectively), in changing from divergent to parallel illumination. That is, multiplying the prefactor in equations (1.5) and (1.6) of Kennedy et al. (2010), which is \((\sqrt{L_M}/\pi)(9\Delta f/(8L_M - 6\Delta f))\), by the change in magnification

\[
\frac{\tilde{M}}{M} = \frac{2/3(1 - 3\Delta f/4L_M)}{2/3(1 - 3\Delta f/8L_M)} = \frac{8L_M - 6\Delta f}{8L_M - 3\Delta f},
\]

gives a prefactor of \((\sqrt{L_M}/\pi)(9\Delta f/(8L_M - 3\Delta f))\), as per equations (3.5) and (3.6) here. Thus, for small objective lens defocus \(\Delta f \ll 4L_M/3\), these shifts are negligibly affected by the change in MEM geometry.

We may express the electron position shifts in terms of the blurred height \(H_B\).

For example, \(S_x\) becomes

\[
S_x(x, y, \delta, \Delta f) = \left( \frac{\partial}{\partial x} \right) H_B(x, y, \delta, \Delta f),
\]

and similarly for \(S_y\), with

\[
H_B(x, y, \delta, \Delta f) = \frac{\Delta f}{8L_M - 3\Delta f} H(x, y) \otimes B(x, y, \delta),
\]

and blurring function

\[
B(x, y, \delta) = \frac{9\sqrt{L_M}}{\pi} (\delta^2 + x^2 + y^2)^{-3/4} (2E_E(x, y, \delta) - E_K(x, y, \delta)).
\]

The shift in electron positions redistributes the intensity on the plane \(z = 4L_M/3 + \Delta f\), which can be derived from electron flux conservation giving (Dyukov et al. 1991; Nepijko et al. 2001b; Kennedy et al. 2010)

\[
I \left( x + \frac{\partial H_B}{\partial x}, y + \frac{\partial H_B}{\partial y}, \delta, \Delta f \right) = \frac{1}{|1 + \frac{\partial^2 H_B}{\partial x^2} + \frac{\partial^2 H_B}{\partial y^2} + (\frac{\partial^2 H_B}{\partial x^2})(\frac{\partial^2 H_B}{\partial y^2}) - (\frac{\partial^2 H_B}{\partial x \partial y})^2|},
\]

in terms of the blurred height \(H_B\).

\[
4. \text{ Laplacian image contrast in mirror electron microscopy with parallel illumination}
\]

We now consider the geometrical theory of MEM contrast in the limit of small objective lens defocus and/or slowly varying \(H(x, y)\). This limit requires that the derivatives of the blurred height are small,

\[
\left| \frac{\partial^2 H_B(x, y, \delta, \Delta f)}{\partial x^2} \right| \ll 1 \quad \text{and} \quad \left| \frac{\partial^2 H_B(x, y, \delta, \Delta f)}{\partial y^2} \right| \ll 1,
\]

which for simplicity, we will refer to as

\[
|\nabla^2_{\perp} H_B(x, y, \delta, \Delta f)| \ll 1,
\]

where \(\nabla^2_{\perp}\) is the transverse Laplacian \((\partial^2/\partial x^2 + \partial^2/\partial y^2)\). The conditions (4.1) and (4.2) are met for a sufficiently slowly varying \(H\) and for defocus that satisfies

\[
|\Delta f| \ll \frac{8L_M}{3 + \max_{x,y} |\nabla^2_{\perp} H(x, y) \otimes B(x, y, \delta)|},
\]

where \( \max_{x,y} g(x, y) \) denotes the maximum value of \( g(x, y) \) over the range of points \((x, y)\). Note that smoothness of the height profile is not required, only that the Laplacian of the height profile (blurred by the function \( B \)) and/or the defocus is small enough to satisfy equations (4.1) and (4.3). In comparison with the equivalent validity condition for diverging illumination, equation (3.3) of Kennedy et al. (2010), noting that the modified form of \( B(x, y, \delta) \) for parallel illumination (equation (3.11)) is twice that of \( B \) for diverging illumination (eqn (2.10) of Kennedy et al. (2010)), under parallel illumination we have a factor of 3 in the denominator of equation (4.3) rather than 6. This suggests that for parallel illumination, Laplacian imaging theory is valid for a broader range of defocus values.

In the limit of small defocus \( \Delta f \) and/or slowly varying \( H(x, y) \) ensuring small derivatives of the blurred height, the intensity (equation (3.12)) is approximated by (Kennedy et al. 2010)

\[
I(x, y, \delta, \Delta f) \approx 1 - \nabla_{\perp}^2 H_B(x, y, \delta, \Delta f).
\]

This is valid for small shifts in electron trajectory, so we have neglected both the change in \( x, y \) coordinates in \( I(x, y, \delta, \Delta f) \) and derivatives greater than second order. Since we have assumed the derivatives are small (equation (4.1)), we have also taken the binomial approximation in the denominator. The blurred height contains the constant term \( \Delta f/(8 L M - 3 \Delta f) \) (equation (3.10)), so for defocus values satisfying \( \Delta f \ll 8 L M/3 \) we have an intensity of

\[
I(x, y, \delta, \Delta f) \approx 1 - \Delta f \nabla_{\perp}^2 H(x, y) \odot \frac{B(x, y, \delta)}{8 L M}.
\]

We note that because \( B(x, y, \delta) \) for parallel illumination (equation (3.11)) is twice that for diverging illumination, the intensity (equation (4.5)) is identical to the intensity expression for divergent illumination, equation (3.7) of Kennedy et al. (2010). Consequently, the simulations and interpretation of droplet trail contrast in §4 and §5 of Kennedy et al. (2010) equally apply to both geometries.

While we consider parallel illumination here, the Laplacian imaging theory can be applied to similar imaging geometries. The general effect will be to multiply the electron shifts with the term \( \bar{M}/M \), where \( \bar{M} \) is the virtual image plane magnification in an existing geometry, and \( M \) is the magnification in the new geometry. Lastly, where these approximations are not valid, due, for example, to either large defocus and/or strong surface height variations, alternative methods such as numerical ray tracing or the recently developed caustic imaging theory may be employed (Kennedy et al. 2011). For fully quantitative simulations, alternative methods may also require a more rigorous treatment of the electric field variations throughout the path of the electron beam, for example, the distortion of the equipotential surfaces in the vicinity of the anode aperture, which was approximated by a thin diverging lens here.

5. Extensions and the inverse problem

Kennedy et al. (2010) discuss a number of extensions, such as the inclusion of chromatic aberration. These extensions are equally applicable to the modern geometry considered here, with the same equations used provided the appropriate
expressions for the blurred height and blurring function are used (equations (3.10) and (3.11), respectively). In particular, to include chromatic aberration, we replace the monochromatic blurring function \( B(x, y, \delta_0, \Delta f) \) with the chromatically averaged \( B_C(x, y, \delta_0, \Delta f) \), given by (Kennedy et al. 2010)

\[
B_C(x, y, \delta_0, \Delta f) = \frac{\sqrt{L - \delta}}{\pi} \left( \frac{9(\Delta f + 2(\delta - \delta_0))}{8(L - \delta) - 3(\Delta f + 2(\delta - \delta_0))} \right) (\delta^2 + x^2 + y^2)^{-3/4} \times (2E_E(x, y, \delta) - E_K(x, y, \delta)) D(\delta) d\delta.
\] (5.1)

Here, a defocus of \( \Delta f + 2(\delta - \delta_0) \) ensures that each intensity corresponds to the plane \( z = 4(L - \delta_0)/3 + \Delta f \), and where \( \delta_0 \) is the mean of the distribution. Chromatic aberration, then, can be incorporated into the Laplacian imaging theory by averaging over several blurring functions to obtain the effective blur \( B_C \). With a normalized distribution we then have (Kennedy et al. 2010)

\[
I_C(x, y, \delta_0, \Delta f) \approx 1 - \nabla^2_{\perp} H(x, y) \ast B_C(x, y, \delta_0, \Delta f).
\] (5.2)

Kennedy et al. (2010) also considers the inverse problem whereby image contrast is analysed to estimate the perturbed electric potential and/or the height variation of the specimen. In the Laplacian imaging theory of MEM contrast this may be achieved using the Fourier derivative theorem (Cowley 1995; Paganin 2006) to convert between spatial derivatives and Fourier space coordinates,

\[
\mathcal{F}(I(x, y, \delta, \Delta f) - 1) \approx \mathcal{F}(-\nabla^2_{\perp} H_B(x, y, \delta, \Delta f)) = (k_x^2 + k_y^2) \mathcal{F}H_B(x, y, \delta, \Delta f),
\] (5.3)

where \( k_x \) and \( k_y \) are the Fourier space coordinates corresponding to real space coordinates \( x \) and \( y \), respectively, \( \mathcal{F} \) is the Fourier transform with respect to \( x \) and \( y \), and \( \mathcal{F}^{-1} \) is the corresponding inverse Fourier transform. This gives (Gureyev & Nugent 1997; Kennedy et al. 2010)

\[
H_B(x, y, \delta, \Delta f) \approx \mathcal{F}^{-1}((k_x^2 + k_y^2)^{-1} \mathcal{F}(I(x, y, \delta, \Delta f) - 1)),
\] (5.4)

namely, the recovery of the blurred height function from a single image, facilitating the analysis of MEM movie dynamics (Tersoff et al. 2009). This expression bears a strong resemblance to phase retrieval via the transport of intensity equation (Teague 1983; Gureyev & Nugent 1997), whereby the original phase object may be recovered via a single phase contrast image.

After obtaining the blurred height function, which depends on the parameters of the MEM, we then deconvolve to obtain the height function, for example via equation (3.10) and using the convolution theorem (Cowley 1995)

\[
H(x, y, \delta) = \frac{(8L_M - 3\Delta f)}{2\pi \Delta f} \mathcal{F}^{-1} \left( \frac{\mathcal{F}(H_B(x, y, \delta, \Delta f))}{\mathcal{F}(B(x, y, \delta))} \right).
\] (5.5)

If the value of the defocus is not known, we can only recover the height to within the scaling factor \( (8L_M - 3\Delta f)/\Delta f \). The scaling factor is the sole difference between equation (5.5) and the equivalent equation (7.3) for divergent
illumination in Kennedy et al. (2010). But since the preliminary examples of a droplet trail in fig. 9 of Kennedy et al. (2010) were recovered to within the scaling factor, the examples are unchanged assuming parallel illumination.

6. Conclusions

We have applied the recently developed Laplacian imaging theory of MEM to an imaging geometry where a converging electron beam is used to ensure parallel illumination of the specimen. Within the domain of validity of the aperture lens approximation and for small defocus, the expressions for the MEM image contrast are unchanged when compared with divergent illumination, and the results and extensions considered by Kennedy et al. (2010) apply. We have shown that the Laplacian imaging theory for parallel illumination has a broader range of valid defocus values. For larger defocus, a scaling factor is geometry dependent, but the Laplacian imaging theory remains a valid and intuitive method of interpreting MEM image contrast.

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