Laplacian image contrast in mirror electron microscopy

BY S. M. KENNEDY, C. X. ZHENG, W. X. TANG, D. M. PAGANIN AND D. E. JESSON*

School of Physics, Monash University, Victoria 3800, Australia

We discuss an intuitive approach to interpreting mirror electron microscopy (MEM) images, whereby image contrast is primarily caused by the Laplacian of small height or potential variations across a sample surface. This variation is blurred slightly to account for the interaction of the electrons with the electrical potential away from the surface. The method is derived from the established geometrical theory of MEM contrast, and whilst it loses quantitative accuracy outside its domain of validity, it retains a simplicity that enables rapid interpretation of MEM images. A strong parallel exists between this method and out of focus contrast in transmission electron microscopy (TEM), which allows a number of extensions to be made, such as including the effects of spherical and chromatic aberration.

Keywords: mirror electron microscopy; Laplacian image contrast; phase contrast; Ga droplets; GaAs

1. Introduction

Mirror electron microscopy (MEM) is a well-established technique that has seen wide application in the real-time study of surface phenomena. Applications include the study of chemical processes at solid surfaces (Świech et al. 1993), surface magnetic fields (Barnett & Nixon 1967a), electric-field contrast (Luk’yanov et al. 1974; Bok 1978; Slezák et al. 2000; Shimakura et al. 2008) and droplet surface dynamics (Tersoff et al. 2009). MEM is unique in surface electron microscopy in that electrons neither impact nor are emitted from the specimen surface. Instead, a near-normally incident beam is reflected at equipotential surfaces just above the specimen. This is achieved by holding the specimen at a small negative voltage relative to the electron source. As the electrons reverse direction, they are travelling very slowly and are consequently sensitive to spatial and/or temporal variations in microfields in the vicinity of the surface. These microfields may, for example, result from small variations in the electric field above the cathode caused by the surface topography (Bauer 1998; Nepijko et al. 2001b; Speake & Trenkel 2003) and/or variations in the electric potential of the specimen itself, including contact potentials, surface charges and varying

*Author for correspondence (david.jesson@sci.monash.edu.au).
conductivity (Barnett & Nixon 1967a,b; Luk’yanov et al. 1974; Bok 1978; Święch et al. 1993; Godehardt 1995). MEM therefore has a significant advantage in that it can probe surface phenomena benignly, without electrons impacting the surface.

The reflected electrons in MEM contain information concerning microfields that are, in turn, related to the topography and/or the electrical and magnetic properties of the surface. This has stimulated significant efforts over the years to interpret MEM image contrast and extract quantitative information regarding the microfields and surface properties. Although a variety of approaches have been employed, including some based on wave mechanics (Hermans & Petterson 1970; Kennedy et al. 2006), most have been based on geometrical ray-tracing techniques (Barnett & Nixon 1967b; Sedov 1970; Luk’yanov et al. 1974; Someya & Kobayashi 1974; Bok 1978; Rempfer & Griffith 1992; Święch et al. 1993; Godehardt 1995).

Building on this latter work, a geometrical theory has been developed in which MEM contrast is viewed as a transverse redistribution of electron current density on an imaging screen due to shifts in electron trajectories following interaction with microfields just above the specimen surface (Sedov 1970; Dyukov et al. 1991; Nepijko & Sedov 1997). This work, which has been used extensively to simulate MEM and photoemission electron microscopy (PEEM) contrast in a variety of situations (Nepijko et al. 2001a,b, 2003, 2007; Jesson et al. 2007; Tang et al. 2009; Nepijko & Schönhense 2010), will form the basis of this paper and we henceforth refer to this approach as the geometrical theory of MEM contrast.

An advantage of the geometrical theory of MEM contrast is that, for special geometries, the electron shifts can be calculated analytically, which can provide important insight into the mechanisms of contrast (Nepijko & Sedov 1997; Nepijko et al. 2001a). Presently, however, there is no direct way of intuitively interpreting MEM contrast of a given general specimen. Here, we present a theory of Laplacian image contrast (see Berry 2006) in MEM that is an approximation of the geometrical theory, yet applicable to a wide range of practical imaging situations. The advantage of the theory is that the image contrast can be interpreted in terms of the Laplacian of an effective two-dimensional phase object that is directly related to the near-surface microfield. For variations in surface topography, the effective phase is related to a blurred-surface height function so that the contrast can be intuitively linked to surface features. Even beyond its strict range of applicability, Laplacian image contrast retains a simplicity that enables rapid interpretation of MEM images. We will show that a strong parallel exists between this method and ‘out of focus’ contrast in transmission electron microscopy (TEM; Cowley 1995; Spence 2003). This allows a number of extensions to be made to the intuitive method, such as including the effects of spherical and chromatic aberration.

2. Geometrical theory of mirror electron microscopy contrast

Dyukov et al. (1991) and Nepijko & Sedov (1997), building upon the earlier work of Sedov (1970), Luk’yanov et al. (1974) and others (Barnett & Nixon 1967b; Bok 1978), have developed a robust geometrical theory of MEM contrast. The approach uses a predominantly classical ‘ray-based’ description of the electron motion inside the imaging system. While the major results are quoted by
A classical electron trajectory (solid lines), travelling parallel to the optical axis $z$ along the centre of an anode aperture $A$, are deflected away from the axis due to the aperture acting as a diverging lens, both upon entering and exiting the anode–cathode region. The aperture separates an electric field free region ($z < 0$) from a constant electric field of $V/L$ ($0 \leq z \leq L$), where the cathode specimen $S$ is held at potential $V < 0$ compared with the anode. An electron of energy $U < -eV$ turns at a distance of $z = L_M$. The $y$-axis extends out of the page. Based on Nepijko & Sedov (1997).

Nepijko & Sedov (1997) and Nepijko et al. (2001b, 2003), and many salient points of the theory are emphasized by Luk'yanov et al. (1974), the foundations of the methodology are less accessible (Dyukov et al. 1991). Since the geometrical theory is the basis for our development of a theory of Laplacian image contrast in MEM, we therefore briefly summarize the key steps here, highlighting the assumptions used in the general case, as well as adapting the method to a low-energy electron microscope (LEEM) imaging system.

A typical electrostatic MEM immersion lens is shown schematically in figure 1. Here, the $z$-axis coincides with the optical axis of the immersion lens and the planar sample surface corresponds to the $(x, y)$-plane of a Cartesian coordinate system. The specimen is held at a negative potential ($V < 0$) relative to the grounded anode aperture a distance $L$ away. The specimen therefore acts as the cathode of the immersion objective lens (Barnett & Nixon 1967b; Luk’yanov et al. 1974; Bok 1978; Bauer 1985). Electrons, accelerated to initial energy $U$, travel along the optic axis, pass through the anode aperture (figure 1) and are deflected by the difference in electric field on either sides of the aperture (Grant & Phillips 1990). For a perfectly smooth sample surface, the electric field between anode and cathode is uniform (except very close to the aperture), and we may trace the electron path classically, whereby the electron moves along a parabolic path as shown in figure 1. If the potential $V$ is chosen such that the electron has zero energy at the cathode surface, i.e. $U = -eV$ with electronic charge $-e$, the classical turning point is at $z = L$. Experimentally, it is customary to adjust $V$ so that $U < -eV$ and the classical turning point is at $z = L_M$ as per figure 1, which is located at a distance of $\delta$ above the specimen surface. For simplicity, deflections in the $y$-direction are not shown, but they are treated independently in the same fashion.

The objective lenses of modern LEEM instruments frequently consist of the electrostatic MEM immersion lens shown in figure 1 combined with a magnetic imaging part (Bauer 1994). To a good approximation, these two components...
can be treated separately (Bauer 1985). As shown in figure 2, the effect of the homogeneous electric field on the trajectory of an electron which turns around a distance $\delta$ above the surface is that it appears to originate from the point $P$, located in the virtual image plane at a distance $2L_M$ from the anode where $L_M = L - \delta$. The effect of the anode aperture is incorporated by assuming that the uniform field is terminated by an ideal diverging lens (Grant & Phillips 1990; Rempfer & Griffith 1992; Nepijko & Sedov 1997), as shown in figure 2. The virtual specimen created by the uniform field at $z = 2L_M$ is the object of the aperture lens with focal length $f = -4L_M$. This lens forms a virtual image of the virtual specimen at point $Q$, which is located in a virtual image plane a distance $4L_M/3$ from the anode. This is the object plane of the magnetic LEEM objective lens.

The geometrical theory of MEM contrast (Dyukov et al. 1991; Nepijko & Sedov 1997) considers the interaction of an electron with variations in the electrical potential $V(x, y, \bar{z})$ above the sample surface, where $\bar{z} = L - z$. This potential is associated with a local surface potential function $V(x, y, \bar{z} = 0)$ that may, for example, arise due to areas of differing work function or applied voltage. A further case arises when the surface is equipotential but varies in height. This situation is equivalent to a planar surface with a corresponding potential distribution (Nepijko & Sedov 1997)

\[
V(x, y, \bar{z} = 0) = \frac{VH(x, y)}{L}, \tag{2.1}
\]

where $H(x, y)$ specifies the surface height of the specimen. In this paper, we will chiefly concentrate on situations of MEM contrast from variations in surface topography via equation (2.1). However, we emphasize that the discussion is entirely valid for variations in potential that can be incorporated directly in
V(x, y, \tilde{z} = 0). By solving the Dirichlet problem for Laplace’s equation for a half space, we have (Polozhiy 1967; Boudjelkha & Diaz 1972; Nepijko & Sedov 1997)

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

(2.2)

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

\[
V(x, y, \tilde{z}) = \frac{\tilde{z}}{2\pi} V(x, y, \tilde{z} = 0) \ast (x^2 + y^2 + \tilde{z}^2)^{-3/2}.
\]

(2.3)

From equation (2.1), 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,

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

(2.4)

Physically, the smoothing function represents the blurring and softening of the electric field when moving away the cathode surface. This smoothed potential will therefore extend beyond the (x, y) range of a localized hill or valley described by H(x, y), for example. The additional potential V(x, y, \tilde{z}) rapidly approaches zero as \tilde{z} increases away from the surface. The geometrical theory therefore assumes that any change to the electron motion caused by the finite height variation of the cathode occurs very close to the sample surface. In addition, the z-dimension motion is assumed to be 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. This amounts to a column approximation, whereby an electron entering the anode at (x_0, y_0) is affected most strongly by the cathode at (3x_0/2, 3y_0/2), where it is closest to the surface (see figure 2). The x and y derivatives of the potential, integrated along the z-axis for the column (3x_0/2, 3y_0/2) therefore give the change to the x and y velocities, respectively. Using the approach of Nepijko & Sedov (1997) and Dyukov et al. (1991), the shift of electron position S_x, S_y on the plane z = 4L_M/3 + \Delta f due to H(x, y) for a small defocus \Delta f of the magnetic objective lens (see figure 3) is given by

\[
S_x(x, y, \delta, \Delta f) = \frac{\partial}{\partial x} \sqrt{\frac{L_M}{\pi}} \frac{9\Delta f}{8L_M - 6\Delta f} H(x, y) \ast ((\delta^2 + x^2 + y^2)^{-3/4} \times (2E_E(x, y, \delta) - E_K(x, y, \delta)))
\]

(2.5)

and

\[
S_y(x, y, \delta, \Delta f) = \frac{\partial}{\partial y} \sqrt{\frac{L_M}{\pi}} \frac{9\Delta f}{8L_M - 6\Delta f} H(x, y) \ast ((\delta^2 + x^2 + y^2)^{-3/4} \times (2E_E(x, y, \delta) - E_K(x, y, \delta)))
\]

(2.6)
Figure 3. The unperturbed (grey line) and perturbed (black line) electron trajectories are traced back along their apparent straight line paths (dashed lines) to the plane $z = 4L_M/3 + \Delta f$. The difference in their position $\Delta r$ is scaled by the expected magnification of the image on this plane relative to the cathode surface $S$, to obtain the electron position shifts $S_x$ and $S_y$ in the specimen plane.

where

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

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

(2.7)

and $K$ and $E$, respectively, denote complete elliptic integrals of the first and second kind (Abramowitz & Stegun 1964; Borwein & Borwein 1987). Here, the magnitude of the electron shift is scaled to the object coordinates (Dyukov et al. 1991; Nepijko & Sedov 1997). Note that for $\Delta f = 0$, the electron shifts are zero, even for a rough surface with non-zero $H(x, y)$. The plane $z = 4L_M/3$ therefore corresponds to the in-focus plane of minimum contrast and a finite defocus $\Delta f$ is required to obtain image contrast. In the special case where $\delta = 0$, the electron has sufficient energy to reach the surface, and the shifts simplify to

$$
S_x(x, y, \Delta f) = \left(\frac{\partial}{\partial x}\right)\sqrt{\frac{L}{\pi^3}}\frac{9\Delta f}{8L - 6\Delta f} \Gamma\left(\frac{3}{4}\right)^2 H(x, y) \otimes (x^2 + y^2)^{-3/4},
$$

(2.8)

and similarly for $S_y$. For later convenience, we separate the derivatives in $S_x$ and $S_y$ from the convolution of the height with the blurring function, introducing the blurred height $H_B$

$$
H_B(x, y, \delta, \Delta f) = \frac{\Delta f}{4L_M - 3\Delta f} H(x, y) \otimes B(x, y, \delta).
$$

(2.9)

The blurring function is

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

(2.10)
which incorporates the smoothing or softening of the electric field as we move away from the cathode surface (see equation (2.4)), and the resulting interaction of the electron with this field. Note that the factor $\Delta f/(4L_M - 3\Delta f)$ in equation (2.9) also contributes to the blurring of the height, but it is kept separate from $B(x, y, \delta)$ for later convenience. Equation (2.5), for example, can then be expressed as

$$S_x(x, y, \delta, \Delta f) = \frac{\partial}{\partial x} H_B(x, y, \delta, \Delta f). \quad (2.11)$$

The shifts in electron position defined by equations (2.5) and (2.6) result in a redistribution of intensity on the plane $z = 4L_M/3 + \Delta f$. The new intensity distribution can be derived from electron-flux conservation, giving (Dyukov et al. 1991; Nepijko et al. 2001b)

$$I(x + S_x, y + S_y) = \frac{I_0(x, y)}{|1 + \partial S_x/\partial x + \partial S_y/\partial y + (\partial S_x/\partial x)(\partial S_y/\partial y) - (\partial S_x/\partial y)(\partial S_y/\partial x)|}, \quad (2.12)$$

where $I_0(x, y)$ is the unperturbed intensity distribution on the plane corresponding to $H(x, y) = 0$ and is typically taken as unity. Intensity values are therefore calculated from the first spatial derivatives of the shift functions, and these are moved from $(x, y)$ to $(x + S_x, y + S_y)$ to evaluate the new intensity distribution.

**3. Laplacian image contrast in mirror electron microscopy**

We now consider the geometrical theory of MEM contrast in the limit of small objective lens defocus and/or slowly varying $H(x, y)$, which is an important practical case frequently encountered in MEM. In addition to the assumptions underpinning the geometrical model highlighted in §2, we require that the derivatives of the blurred height are small,

$$\left|\frac{\partial^2 H_B(x, y, \delta, \Delta f)}{\partial x^2}\right| \ll 1 \quad \text{and} \quad \left|\frac{\partial^2 H_B(x, y, \delta, \Delta f)}{\partial y^2}\right| \ll 1, \quad (3.1)$$

which for simplicity we will refer to as

$$|\nabla^2_\perp H_B(x, y, \delta, \Delta f)| \ll 1, \quad (3.2)$$

where $\nabla^2_\perp$ is the transverse Laplacian $(\partial^2/\partial x^2 + \partial^2/\partial y^2)$. For a given blurring function $B$ (equation (2.10)) that is determined by the experimental parameters, the required limits of equations (3.1) and (3.2) are met with a sufficiently small objective lens defocus $\Delta f$ satisfying

$$|\Delta f| < \frac{4L_M}{(3 + \max_{x,y} |\nabla^2_\perp H(x, y) \otimes B(x, y, \delta)|)}, \quad (3.3)$$

where $\max_{x,y} g(x, y)$ denotes the maximum value of $g(x, y)$ over the range of points $(x, y)$. Conversely, if we require that the maximum $|\Delta f|$ used in a through-focal series of images is large enough to provide significant image contrast, i.e. $|\Delta f| > \alpha$ for some distance $\alpha$, equation (3.3) demands that $H(x, y)$ be sufficiently
slowly varying to satisfy \( \max_{x,y} |\nabla^2 H(x, y) \otimes B(x, y, \delta)| < -3 + 4L_M/\alpha \). 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 (3.1) and (3.2).}

Inserting equations (2.9) and (2.11) into equation (2.12), the image intensity can be expressed in terms of the blurred height function as

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

For small defocus \( \Delta f \) and/or slowly varying \( H(x, y) \) ensuring small derivatives of the blurred height (equations (3.1) and (3.2)), the intensity expression is approximated by

\[
I(x, y, \delta, \Delta f) \approx \frac{1}{|1 + \partial^2 H_B/\partial x^2 + \partial^2 H_B/\partial y^2|}.
\]

This is valid for small shifts in electron trajectory (see equation (2.11)), so that we have neglected the change in \( x, y \) coordinates in \( I(x, y, \delta, \Delta f) \) and derivatives greater than second order. Since the second derivatives in equation (3.5) are much smaller than unity, the denominator will always be positive, so we may remove the absolute value signs and take the binomial approximation of the denominator giving

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

The blurred height contains the constant term \( \Delta f / (4L_M - 3\Delta f) \) (see equation (2.9)), and provided we choose a defocus much smaller than the sample-to-anode distance \( L \), e.g. \( \Delta f = 10^{-5} \text{ m}, L = 10^{-3} \text{ m} \), this term is approximately proportional to the defocus \( \Delta f \). So we may write the intensity as

\[
I(x, y, \delta, \Delta f) \approx 1 - \Delta f \nabla^2 H(x, y) \otimes \frac{B(x, y, \delta)}{4L_M},
\]

where the blurring function \( B(x, y, \delta) \) is given in equation (2.10). This indicates that where the height variation and/or defocus is small enough to satisfy equation (3.2), the image intensity on the ‘out of focus’ plane \( z = 4L_M/3 + \Delta f \) is the Laplacian image of the height function, blurred with a function \( B(x, y, \delta)/4L_M \) to account for the interaction of the electron with the electric field above the cathode surface. In the regime where this approximate expression is valid, we may therefore interpret MEM image contrast to be created solely by the transverse second derivatives (curvature) of the surface height variation, smoothed by a blurring function. This is an important result for the intuitive interpretation of MEM contrast of surface topography.

Laplacian imaging is widely encountered in many contexts ranging from X-ray imaging (Paganin 2006) to oriental magic mirrors (Berry 2006) and their modern equivalent in Makyoh topography (Riesz 2000). It is also known as out of focus
contrast in TEM of thin specimens (Lynch et al. 1975; Cowley 1995; Spence 2003). The applicability of the Laplacian imaging formalism to MEM under particular conditions considerably simplifies image interpretation, as we will discuss in §4.

4. Intuitive interpretation of mirror electron microscopy image contrast

As an application of Laplacian imaging in MEM, we apply the technique to investigate Ga droplets on GaAs (001). This system is known to exhibit droplet surface dynamics that obey an unusual temperature dependence (Tersoff et al. 2009). As Ga droplets move on the rough GaAs (001) surface, they leave behind smooth trails as shown in the atomic force microscope (AFM) image in figure 4. Outside of the trail there is significant surface roughness and we obtain a mean trail profile by averaging the surface height along the y-axis in the framed region shown in figure 4. The resulting averaged cross-sectional profile, contained in figure 5a, is 1.9 μm wide and 14 nm deep. For the range of droplet sizes studied by AFM, we find that the width to depth ratio of the trails is approximately constant (approx. 140). With $L = 2$ mm, $\delta = 40$ nm, $V = -20000.4$ V and $U = 20$ keV, and for the droplet trails considered here, we find that $\max_{x,y} |\nabla^2 H(x, y) \otimes B(x, y, \delta)| \approx 35$ m$^{-1}$ or lower, so that the condition of equation (3.3) requires that $|\Delta f| < 200$ μm in order to satisfy $|\nabla^2 H_B| \ll 1$. Therefore, the assumptions underpinning a Laplacian contrast interpretation as outlined in §3 are valid and we choose the droplet trails as convenient test objects for Laplacian MEM imaging. Note that the height of the droplet itself (denoted ‘D’ in figure 4) is too large (0.3 μm above the cathode surface) to satisfy the assumption that changes in the z-component of the electron motion can be neglected. Therefore, it is inappropriate to apply the geometrical theory and a Laplacian interpretation in this case.

It is experimentally impractical to obtain both AFM and MEM images of the same droplet trail, therefore we consider only the general features of the AFM data of figure 4. Specifically, we ignore the significant surface roughness outside the trail, still present due to the limited area available for averaging, which will inevitably lead to strong intensity fluctuations in MEM images. So rather than use the AFM data directly in the Laplacian MEM method, in this example, we instead model the trail using a height function $H(x)$ that is the sum of two inverse tangent functions,

$$H(x) = \frac{T}{\pi} \left( \tan^{-1} \left( \frac{x - R}{O} \right) - \tan^{-1} \left( \frac{x + R}{O} \right) \right).$$ (4.1)

Here, $T$ sets the maximum depth of the trail, $R$ is the distance of the side from the centre and $O$ sets the steepness of the trail edge, e.g. for $O = 0.1$ μm, 80 per cent of the variation of the trail edge about its midpoint occurs over a distance of 0.5 μm (see figure 5a). A background linear variation in $x$ in the AFM data was ignored when fitting the height function (the variation was removed to give figure 5a), as we consider only the general features of the AFM data in this example. Note that the Laplacian contrast method is insensitive to linear variations in $x$ that span the entire AFM image, since the second derivative of the height dominates the image contrast. However, a linear variation that begins and/or ends within

Figure 4. Atomic force microscope (AFM) image of a trail left by a moving Ga droplet marked D on a GaAs (001) surface. The region inside the box is integrated along y to obtain a one-dimensional height profile in x, shown in figure 5a (scale bar, 1μm).

Figure 5. (a) Averaged one-dimensional profile of a droplet trail on the cathode surface (grey line), along with the simplified height function $H(x)$ (black line) fitted using equation (4.1) with $R = 0.95\,\mu m$, $O = 0.1\,\mu m$, $T = 15\,nm$. (b) Second spatial derivative of $H(x)$ that provides the key qualitative features of the MEM image.

the data range will introduce a discontinuity where the linear variation starts and/or finishes, which has a non-zero second derivative and will contribute to the image intensity.

Fitting equation (4.1) to the general features of the averaged cross-sectional profile gives a simplified model of the trail-height function (see figure 5a). As indicated in figure 5a, we choose a broad trail edge to account for the width variation and surface roughness evident in figure 4. As discussed earlier, a major advantage of Laplacian imaging contrast is its ease of interpretation via equation (3.7). It is therefore straightforward to predict the general features of the image contrast of a droplet trail from the second derivative of the model trail-height function contained in figure 5a. This is shown in figure 5b and indicates that the MEM image should contain a bright and dark contrast band in the vicinity of the trail edges, along with constant intensity in the centre of the trail. We emphasize that such a first-order interpretation of MEM contrast in terms of surface curvature is quite general and independent of the surface profile, provided the Laplacian imaging theory is valid. This has important practical value for studies of surface phenomena using MEM.
Figure 6. (a) MEM image of a moving Ga droplet D and the trail left on a GaAs (001) surface. Imaging conditions were $V = -20000.4 \text{ V}$, $U = 20 \text{ keV}$ and $L = 0.002 \text{ m}$, giving $\delta = 40 \text{ nm}$. Comparison of MEM images and simulations using equation (3.7) of the trail region contained in the frame in (a) are shown for (b) negative defocus ($\Delta f = -15 \mu \text{m}$), (c) approximately zero defocus and (d) positive defocus ($\Delta f = 15 \mu \text{m}$). The trail-height function was approximated using equation (4.1) for $R = 0.83 \mu \text{m}$, $O = 0.1 \mu \text{m}$, $T = 13 \text{ nm}$ (scale bars (a) 2 $\mu$ m and (b) 1 $\mu$ m).

In practice, equation (3.7) indicates that the second derivative of $H$ is softened or smoothed by convolution with the blurring function $B(x, y, \delta)$ in forming the image, physically accounting for the electron interacting with the electric field above the cathode. The defocus $\Delta f$ will affect both the magnitude and the sign of the contrast peaks. A qualitative comparison of simulated Laplacian contrast images, based on equation (3.7), with experimental MEM images of a trail similar to that in figure 4 is shown in figure 6 for negative, zero and positive defocus values. Although the surface roughness outside the trail region results in significant contrast fluctuations, it can be seen that the main features of the experimental image through-focus sequence are consistent with Laplacian imaging theory for a generalized trail profile. A more complex or realistic height profile, e.g. that recovered in §7, can account for image features caused by surface roughness. Figure 7 compares simulations and experimental profiles of the MEM image intensity for positive and negative defocus values. The latter profiles have been integrated over the two-dimensional panel region in figure 6, parallel to the trail edges, to reduce the intensity fluctuations caused by the surface roughness.
Figure 7. Comparison of simulated Laplacian contrast images (black lines) with experimental MEM intensity profiles of a droplet trail (grey lines). The experimental MEM intensity profiles were obtained by spatially averaging the intensities parallel to the trail edge over the two-dimensional regions in figure 6b,d. (a) $\Delta f = -15 \mu m$, (b) $\Delta f = 15 \mu m$. The trail-height function was approximated using equation (4.1) for $R=0.83 \mu m$, $O=0.1 \mu m$, $T=13 nm$. The grey-scale intensity values in the experimental images were scaled to match the vertical axis of the simulations, allowing a qualitative comparison.

roughness. The good agreement in both cases again illustrates the applicability of Laplacian imaging that facilitates the interpretation of image contrast in terms of surface curvature.

5. Comparison of the Laplacian and geometrical theory

It is important to establish and confirm the domain of validity of Laplacian imaging theory. We therefore compare image simulations based on the height profile of the droplet trail shown in figure 5a, using the geometrical (equation (3.4)) and the approximate Laplacian contrast approaches (equation (3.7)). As shown in figure 8a for defocus $\Delta f = -15 \mu m$ and classical turning point $\delta = 40 nm$ from the cathode surface, the two methods agree very closely. Increasing the magnitude of the defocus and/or decreasing the turning-point distance will increase the blurred height $H_B$ and its derivatives. This weakens the validity of the assumption made in the Laplacian contrast method that $|\nabla^2 H_B| \ll 1$, and we therefore see an increased discrepancy between the image contrast generated from the Laplacian contrast and geometrical imaging simulation methods (figure 8b). Conversely, reducing the magnitude of the defocus and/or increasing the turning distance improves the agreement between the two approaches as expected.

6. Extensions of the Laplacian imaging theory of mirror electron microscopy contrast

Having established the applicability of Laplacian imaging theory to MEM we now use previous studies to extend our analysis. In particular, Laplacian contrast is also known as out of focus contrast in TEM of thin specimens (Lynch et al. 1975;
Laplacian image contrast in MEM

Figure 8. Comparison of the one-dimensional intensity profile predicted using the geometrical treatment (grey line) with the Laplacian contrast method (black line), for the droplet trail-height profile of figure 5, using $R = 0.83 \mu m$, $O = 0.1 \mu m$, $T = 13 nm$. (a) $\Delta f = -15 \mu m$, $\delta = 40 nm$; (b) $\Delta f = -30 \mu m$, $\delta = 20 nm$.

Cowley 1995; Spence 2003), and we can use this formalism to include the effects of spherical and chromatic aberration. These aberrations are an intrinsic part of an MEM imaging system and limit resolution (Rempfer & Griffith 1992). Since a Laplacian contrast interpretation is applicable to imaging objects at high resolution provided $|\nabla^2 B| \ll 1$, it is important to incorporate such effects into the imaging theory. The expression for TEM out of focus contrast for a thin uniformly illustrated specimen is (Lynch et al. 1975; Cowley 1995; Spence 2003)

$$I(x, y, z = z_0 + \Delta f) = 1 - k^{-1} \Delta f |\nabla^2 \phi|,$$  

(6.1)

for a defocus $\Delta f$ and electron wavenumber $k = 2\pi/\lambda$. The electron phase change through the specimen $\phi$ is inversely proportional to the local electron wavelength $\lambda$ so that the wavelength dependence factors out in equation (6.1) and so it is possible to extrapolate the wavelength to zero (cf. equation (6.4)).

We note that equation (6.1) is identical to the Laplacian theory description of MEM contrast (equation (3.7)) provided the phase of the wave function is

$$\phi(x, y, \delta) = \frac{k}{\Delta f} H_B(x, y, \delta, \Delta f).$$  

(6.2)

We may view this as the effective phase variation of an electron wave post interaction with the cathode sample surface, which has been scaled up to the vacuum or post anode aperture energy. Equation (6.1) therefore describes the out of focus MEM contrast in the defocused image plane $z = 4L_M/3 + \Delta f$.

Lynch et al. (1975) extended the TEM out of focus expression to include the effects of spherical aberration, which depends on the bi-Laplacian or iterated Laplacian ($\nabla^4 \equiv \nabla^2 \nabla^2 \nabla^2$) of the phase variation $\phi$, scaled by the spherical aberration coefficient $C_S$,

$$I(x, y, \delta, \Delta f) \approx 1 - \frac{\Delta f}{k} |\nabla^2 \phi(x, y, \delta)| + \frac{C_S}{2k^3} \nabla^4 \phi(x, y, \delta).$$  

(6.3)
We may recast this equation using equation (6.2) to give

\[ I(x, y, \delta, \Delta f) \approx 1 - \nabla^2 H_B(x, y, \delta, \Delta f) + \frac{C_s}{2\Delta f^2} \nabla^4 H_B(x, y, \delta, \Delta f), \]  

(6.4)

which extends our Laplacian contrast expression to include spherical aberration. For the resolutions employed in the study of droplet trails and with \(C_s\) values derived by Rempfer & Griffith (1992), we have found that including spherical aberration provides less than a 1 per cent change in the simulated intensity variation. However, we anticipate that the inclusion of spherical aberration will be of benefit in simulating higher resolution images of surface objects within the domain of validity of Laplacian imaging.

We may also extend the Laplacian contrast method to include the effects of a finite energy spread in the electron beam, which causes chromatic aberration in the image intensity. A distribution in energy \(D(U)\) varies the classical turning point \(\delta\), via

\[ \delta = L \left( 1 + \frac{U}{eV} \right), \]  

(6.5)

where the cathode surface is kept at a potential of \(V < 0\). The distribution in turning point \(D(\delta)\) can then be obtained from the energy distribution, e.g. \(D(\delta) \approx D(U) \, dU/d\delta\). Following the approach of Fejes (1977), we incoherently average over the distribution, summing up the contributions of each intensity (equation (3.7)) weighted by the distribution function

\[ I_C(x, y, \Delta f) = \int I(x, y, \delta, \Delta f)D(\delta) \, d\delta \approx \int D(\delta) \, d\delta - \nabla^2 \int H_B(x, y, \delta, \Delta f)D(\delta) \, d\delta. \]  

(6.6)

Since the turning distance \(\delta\) only appears in the blurring function, in effect we may replace the monochromatic blurring function \((\Delta f/(4L_M - 3\Delta f))B(x, y, \delta)\) with the chromatically averaged \(B_C(x, y, \delta_0, \Delta f)\) given by

\[ B_C(x, y, \delta_0, \Delta f) = \int \frac{\sqrt{L - \delta}}{\pi} \frac{9(\Delta f + 2(\delta - \delta_0))}{8(L - \delta) - 6(\Delta f + 2(\delta - \delta_0))} (\delta^2 + x^2 + y^2)^{-3/4} \]

\[ \times (2E_E(x, y, \delta) - E_K(x, y, \delta))D(\delta) \, d\delta, \]  

(6.7)

with a defocus of \(\Delta f + 2(\delta - \delta_0)\) to ensure 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 approximate method by adjusting the blurring function, in essence averaging over several blurring functions to obtain the effective blurring function \(B_C\). With a normalized distribution, we then have

\[ I_C(x, y, \delta_0, \Delta f) \approx 1 - \nabla^2_H(x, y) \otimes B_C(x, y, \delta_0, \Delta f). \]  

(6.8)

As with spherical aberration, chromatic aberration has a small effect on simulating the MEM image contrast of the droplet trails (less than 1% as expected). This is true for a Gaussian energy distribution with a typical full-width-half-maximum equal to 0.3 eV for a Schottky field-emission source and a variety of mean \(\delta_0\) values. However, we would again envisage that equation (6.8) will be of value for the study of surface objects at high resolution within the Laplacian imaging regime of \(|\nabla^2 H_B| \ll 1\).
7. Inverse problem of Laplacian mirror electron microscopy imaging

Many of the geometrical treatments consider the important ‘inverse problem’ of MEM imaging, whereby image contrast is analysed to estimate the perturbed electric potential and/or the height variation of the specimen (Luk’yanyov et al. 1974; Dyukov et al. 1991; Nepijko & Sedov 1997; Nepijko & Schönhense 2010). The inverse problem has also been explored in other areas of surface electron microscopy such as LEEM (Yu et al. 2010). In the Laplacian theory of MEM contrast, this may be achieved in a very straightforward fashion using the Fourier derivative theorem (Cowley 1995; Paganin 2006) to convert between spatial derivatives and Fourier-space coordinates,

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

(7.1)

Here, \(k_x\) 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. We therefore have (Gureyev & Nugent 1997)

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

(7.2)

which in principle allows 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; Paganin & Nugent 1998), whereby a phase contrast image may be used to recover the original phase object.

Upon obtaining the blurred height function, we then deconvolve to obtain the height function, for example via equation (2.9) using the convolution theorem (Cowley 1995)

\[
H(x, y, \delta) = \frac{(4L_M - 3\Delta f)}{2\pi\Delta f} \mathcal{F}^{-1} \left( \frac{\mathcal{F}(H_B(x, y, \delta, \Delta f))}{\mathcal{F}(B(x, y, \delta))} \right).
\]

(7.3)

If the value of the defocus is not known, we can only recover the height to within the scaling factor \((4L_M - 3\Delta f)/\Delta f\). Here, we present two preliminary examples in one dimension of the inverse problem of Laplacian MEM imaging. Figure 9a shows the recovered height using equations (7.2) and (7.3) from the simulated MEM images shown in figure 7. The recovered height is in very good agreement with the ideal height profile of equation (4.1), also shown in figure 9a.

Figure 9b shows an average of the recovered heights from the experimental MEM intensity profiles of figure 7 to within a scaling factor, as the specific defocus values were not known. The general features of the recovered height are in good agreement with the ideal height profile, with discrepancies largely due to the surface roughness evident in the recovered height profile. Using equation (3.7) to simulate the Laplacian image contrast of the recovered height profile of figure 9b, we found that the normalized root-mean-squared difference between the simulated intensity and the measured MEM image intensity profiles (figure 7) was 3 and 10 per cent for the negative and positive defocus images, respectively.
Figure 9. Recovered height profiles of the droplet trail (black lines) using equations (7.2) and (7.3) compared with the ideal height profile (grey lines) of equation (4.1) with $R = 0.83\, \mu m$, $O = 0.1\, \mu m$, $T = 13\, nm$. (a) The recovered height from the simulated MEM intensity profiles for the ideal height (black lines in figure 7), using $\Delta f = -15\, \mu m$. (b) Average of the recovered height profiles (black line) of the experimental MEM images (grey lines in figure 7). The recovered height $H(x, \Delta f) = H(x)\Delta f/(4L_M - 3\Delta f)$ includes the scaling factor $\Delta f/(4L_M - 3\Delta f)$ since $\Delta f$ in each image was unknown.

8. Conclusions

We have demonstrated that Laplacian imaging theory can be applied to MEM imaging of surface topography (or equivalently surface potentials) provided the height function describing the surface topography is slowly spatially varying and/or the objective lens defocus is small. Under such conditions, image contrast is primarily caused by the Laplacian of small height or potential variations across a sample surface. This contrast is blurred due to the interaction of the electrons with the electrical potential away from the surface. However, the method facilitates the rapid and intuitive interpretation of image contrast in terms of surface topographic or potential variations. The approach can be readily extended to include spherical and chromatic aberration. Finally, we have demonstrated that the Laplacian imaging theory forms a convenient basis for the solution of the inverse problem in MEM.

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References


Laplacian image contrast in MEM


Addendum. Laplacian image contrast in mirror electron microscopy

BY S. M. KENNEDY, C. X. ZHENG, W. X. TANG, D. M. PAGANIN AND D. E. JESSON*

School of Physics, Monash University, Victoria 3800, Australia

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.

Keywords: mirror electron microscopy; Laplacian image contrast; phase contrast

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.

*Author for correspondence (david.jesson@monash.edu).
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’yakov et al. (1974), Dyukov et al. (1991), Godheardt (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’yakov 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 $H(x, y)$,
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 \(z\) at \(z = 4L_M\). 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; Rempfer & 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 $V(x, y, \tilde{z})$ rapidly approaches zero as $\tilde{z}$ 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

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

$$
\times (2E_E(x, y, \delta) - E_K(x, y, \delta)) \tag{3.5}
$$

and

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

$$
\times (2E_E(x, y, \delta) - E_K(x, y, \delta)), \tag{3.6}
$$

where

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

and

$$
E_K(x, y, \delta) = K \left( \frac{1}{2} - \frac{\delta}{2(\delta^2 + x^2 + y^2)^{1/2}} \right), \tag{3.7}
$$

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

$$
\frac{\tilde{M}}{M} = \frac{2/3(1 - 3\Delta f/4L_M)}{2/3(1 - 3\Delta f/8L_M)} = \frac{8L_M - 6\Delta f}{8L_M - 3\Delta f}, \tag{3.8}
$$

gives a prefactor of \((\sqrt{T_M}/\pi)(9\Delta f/(8L_M - 3\Delta f))\), 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

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

and blurring function

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

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)

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

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

4. 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

\[
|\nabla^2 T H_B(x, y, \delta, \Delta f)| \ll 1,
\]

where \(\nabla^2 T\) 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

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

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)

\[
I(x, y, \delta, \Delta f) \approx 1 - \nabla^2 H_B(x, y, \delta, \Delta f). \tag{4.4}
\]

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 \( \Delta f / (8L_M - 3\Delta f) \) (equation (3.10)), so for defocus values satisfying \( \Delta f \ll 8L_M/3 \) we have an intensity of

\[
I(x, y, \delta, \Delta f) \approx 1 - \Delta f \nabla^2 H(x, y) \ast \frac{B(x, y, \delta)}{8L_M}. \tag{4.5}
\]

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 \((\Delta f/(8L_M - 3\Delta f))B(x, y, \delta)\) with the chromatically averaged \(B_C(x, y, \delta_0, \Delta f)\), given by (Kennedy et al. 2010)

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

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)

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

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,

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

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)

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

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)

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

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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References


Addendum. Laplacian image contrast in MEM


