Linear imaging of strong phase objects using asymmetrical detectors in STEM

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Received 5 February 1979

Abstract

In transmission electron microscopy it is highly desirable that the image intensity and phase of the specimen transmittance should be linearly related. However, for normal bright field imaging conditions this requires that severe restrictions be imposed on the phase. By using a STEM equipped with detectors which have asymmetrical response functions the severity of these restrictions can be much reduced. The extent to which certain configurations approximate to linear imaging systems for strong phase objects, together with their linear transfer properties, are discussed.

Inhalt


1. Introduction

In transmission electron microscopy the electron wavefunctions just before and beyond the specimen are frequently related by a multiplicative complex function. This function, known as the transmittance, is characteristic of the specimen, but independent of the nature of the wavefunction just before the specimen, at least for all wavefunctions of interest. The validity of this description is restricted, therefore, to cases where diffraction effects within the specimen itself are negligible, and under these conditions the phase of the

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transmittance relates directly to the electrostatic scalar or magnetic vector potential of the specimen.

For weak phase objects, or weak mixed amplitude and phase objects, the transmittance may be expanded to include only first order terms and the resulting expressions for the image contrast are linear, if normal bright field imaging conditions are used. However, such a first order approximation is frequently invalid. Some examples of the violation of weak modulation conditions include the following: (i) small clusters of heavy atoms, as in negatively stained specimens [1] (ii) appreciable abrupt changes in specimen thickness e.g. voids in a specimen or small crystals on a support film [2] (iii) domain walls in ferromagnetic thin films [3]. One aspect of the failure of the first order theory is the production of pseudo amplitude contrast [4], but a more serious effect concerns the complications introduced into image interpretation e.g. non-linear phase imaging.

For such non-linear imaging conditions no closed form relationship between image contrast and the phase function exists. Two approaches to this problem are possible. The first is the classic method of science, that is, a model object is assumed and a suitable theory is used to predict the image; comparison with the experimental image then gives an indication of the validity of the model. A problem with this is that the ambiguities of the observation remain implicit and considerable computing is frequently necessary. A more ambitious plan is to try to solve the non-linear phase problem as is discussed in detail by Saxton [5]. Although the solution of this problem is sometimes feasible the investment in hardware, software and effort is high and it seems unlikely that simple standard techniques will evolve.

Methods for the linear imaging of strong phase or mixed objects are, therefore, of interest. It is the intent of this paper to demonstrate that approximations similar to those used for weak phase or mixed objects in normal bright field microscopy are not restricted to such objects when other imaging systems are employed. Several such systems are considered and the extent to which they approximate to linear imaging systems, together with their linear transfer characteristics, are discussed in the following sections.

2. A characteristic of systems capable of imaging linearly strong phase objects

When considering the possibility of imaging linearly an object which strongly modulates the electron beam, it is constructive, as will become apparent later, to consider a STEM with a detector response function \( R(k) \). The response function is defined by noting that unit current density in the detector plane in the region \( k \) to \( k + dk \) will produce a signal of \( R(k)dk \). Normal STEM geometry is assumed so that the complex disturbance in the detector plane \( \Psi(k, r_0) \) is related to the complex disturbance in the specimen exit plane \( \Psi(r, r_0) \) by a Fourier transform, apart from a familiar scaling and an irrelevant phase factor.
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$$\mathcal{P}(k, r_0) = \mathcal{F} \{ \psi(r, r_0), r \rightarrow k \} = \int_{-\infty}^{\infty} \psi(r, r_0) \exp(-2\pi i k \cdot r) \, dr$$ (1)

Here \(r\) is a position vector in the object plane, \(r_0\) is the position vector of the probe and \(k\) is most conveniently taken to be a spatial frequency vector which is directly proportional to the position vector in the detector plane.

In the absence of any specimen interaction, the probe wavefunction in the object plane is \(\psi_0(r \rightarrow r_0)\), where isoplanatic optics have been assumed, and, because it adds little to the present discussion, partial coherence in the probe is neglected. The specimen interaction is described by a complex transmittance, \(h(r) = a(r) \exp(i\phi(r))\) so that

$$\psi(r, r_0) = h(r) \psi_0(r \rightarrow r_0)$$ (2)

The image signal \(s(r_0)\), which is simply the current falling on the detector when the probe is centred about \(r_0\), may now be written as

$$s(r_0) = \int \mathcal{F} \{ \mathcal{H}(k), r \} \, dk$$

$$= \mathcal{G}(r) \ast [h(r) \psi_0(r \rightarrow r_0)] \ast [h^*(r) \psi_0^*(r \rightarrow r_0)] \bigg|_{r=0}$$ (3)

where \(\ast\) denotes the convolution integral and the subscript \(r = 0\) implies that the expression has to be evaluated at \(r = 0\). The function \(\mathcal{G}(r)\) is defined by

$$\mathcal{G}(r) = \mathcal{F}^{-1} \{ \mathcal{H}(k), k \rightarrow r \}$$ (4)

and is related by the principle of equivalence to an unnormalised form of the degree of coherence [6].

If attention is restricted to pure phase objects, eq. (3) can be written explicitly as

$$s(r_0) = \int \mathcal{G}(r') \psi_0(r' \rightarrow r_0) \psi_0^*(r' \rightarrow r_0) \exp[i\phi(r')] \bigg|_{r=0}$$

$$- i\phi(r' \rightarrow r') \bigg|_{r=0}$$ (5)

Equation (5) is in general non-linear in \(\phi\), although a linear approximation results if it is valid to expand the exponential to include only first order terms. This, however, does not imply that the phase function itself is restricted to a small excursion, for it may be that the function \(\mathcal{G}(r') \psi_0(r' \rightarrow r_0) \psi_0^*(r' \rightarrow r_0)\) has appreciable amplitude only over a finite range of \(r'\). Under these circumstances the contributions to the signal in eq. (5) come predominantly from this region and it is sufficient that the difference \(\Delta\phi = \phi(r') - \phi(r' \rightarrow r')\) be small over this range. As the width of the function \(\mathcal{G}(r)\) is controllable by suitable choice of \(R(k)\) this would seem a possible approach to devising systems suitable for imaging linearly strong phase objects. It should be pointed out, however, that normal phase contrast techniques employ a detector for which \(R(k)\) is small, corresponding to a high equivalent degree of coherence, so that \(\mathcal{G}(r)\) is generally a broad function. How this apparent dilemma may be resolved is discussed in the next section.
Finally, in this section, we consider how the success of a particular system for the linear imaging of strong phase objects may be assessed. If the phase of the object transmittance varies periodically, the Fourier transform of the phase function (\( \Phi(k) \)) will consist of an array of \( \delta \)-functions at integral multiples of the fundamental spatial frequency. Two effects of a non-linear imaging system are the introduction of harmonic distortion and frequency mixing. To investigate such occurrences it is convenient to consider the Fourier transform of the image, which we denote by \( S(k) \). This may be obtained from eq. (3), and after some manipulation we obtain

\[
S(k) = \int \int R(k') H(k' - k') \Psi_0(k - k') \Psi^*_0(k') dk' dk'' \quad (6)
\]

where \( H(k) = \mathcal{F} \{ h(r), r \rightarrow k \} \). In addition to looking for contributions to \( S(k) \) at spatial frequencies absent from \( \Phi(k) \) it is convenient to use this expression to investigate the effect of multiplying the phase function by a constant \( c \). For a linear system, \( S(k) \) should merely scale by the same factor \( c \), a result which is, of course, applicable when isolated as well as periodic objects are of interest. In the following section the range of phase modulation over which the above criteria are approximately true is considered by evaluating eq. (5) and (6) for various test objects and detector response functions.

3. Some examples of detector systems with narrow \( \rho(r) \) functions

3.1 Axial disc detector. As a first example we consider a STEM with a uniform circular detector (semi-angle, \( \beta \)) operating in the brightfield Fresnel or defocus contrast mode. Such calculations are equally valid for a CTEM with a uniform source which subtends the same semiangle, \( \beta \). As a test object we consider initially the phase function defined in fig. 1. Such a phase function may be regarded as an idealisation of that resulting from a small cube (of, for example, magnesium oxide \([7]\)) viewed along a [110] direction or from a zero-width domain wall in a ferromagnetic thin film. In the first example the parameter \( \kappa \) is proportional to the mean inner potential of the crystal, whilst for the latter, \( \kappa \) is proportional to the product of the in-plane induction and foil thickness. Typical values of \( \kappa \) using specimens suitable for investigation with 100 kV electrons are \( \sim 10^9 \) radian m\(^{-2}\) for both the cube and the magnetic sample.

To evaluate eqs. (5) and (6) for the model shown in fig. 1 a computer programme was used which assumed temporal coherence, but which allowed for aberrations in the probe as well as the required variation of \( \beta \) and defocus. At least one specific case, however, can be evaluated analytically in a straightforward manner. This is the case for an unaberrated probe and a detector response function \( R(k) = S(k) \), and corresponds to illuminating the specimen with a plane wave in a CTEM and forming the image with a perfect optical system. Under these conditions, eq. (6) yields (in 1 dimension)
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\[ S(k) = \delta(k) + \gamma \kappa \pi^2 \text{sinc}(\gamma k(k - \kappa^{-1})), \text{ for } ky > 0 \]

\[ = \delta(k) + \gamma \kappa \pi^2 \text{sinc}(\gamma k(k + \kappa^{-1})), \text{ for } ky < 0 \]

where \( \text{sinc}(p) = \sin(p)/p \) and \( \gamma = \pi \lambda Z \), \( \lambda \) and \( Z \) being the electron wavelength and defocus respectively. In general the effect of multiplying \( \kappa \) by a constant \( c \) will be to change markedly the shape of \( S(k) \) indicating the non-linearity of the system. However, if \( \kappa^2 \ll \pi^2 \gamma^{-1} \), \( \text{sinc}(\gamma k(k - \kappa^{-1})) \) will not vary markedly with \( \kappa \) and one criterion for linearity described above will be satisfied approximately. For a magnetic foil with \( \kappa \sim 10^8 \text{ radian m}^{-1} \) the above inequality implies use of defocus distances \( < 25 \mu m \), a factor of \( 10^2 \) lower than those frequently used in Lorentz microscopy. Under these circumstances the contrast in the image would be unacceptably low, emphasizing the inadequacy of conventional phase contrast techniques when confronted with certain strong phase objects of physical interest.

Figure 2 shows the result of evaluating eq. 5 for a pair of objects of the type shown in fig. 1, one having a positive and the other a negative value of the same magnitude for \( \kappa \). \( y \) is held constant and image intensity profiles are

\[ \varphi = \kappa |x| \]

\[ \frac{d\varphi}{dx} = \kappa \text{sgn}|x| \]

**Fig. 1.** Spatial variation of phase and phase gradient of the test object considered.
plotted for 10 values of $\beta$. Alternatively, it should be noted that each profile may be regarded as arising from an object with a positive value of $\kappa$ but with the left hand side of the profile representing the image intensity obtained using a negative $\gamma$ and the right hand side that with a positive $\gamma$ of the same magnitude. As Fresnel contrast responds neither to a constant phase nor to a constant phase gradient the image contrast obtained on reversing the sign of the defocus should be identical apart from a sign reversal, providing the image formation is linear. In accordance with the preceding paragraph, at low $\beta$ the image formation is found to be highly non-linear, but as the figure shows, as $\beta$ increases the non-linearities are reduced. Unfortunately the contrast itself is also reduced because of the lower equivalent degree of coherence and so straightforward attempts at strong phase linear imaging by using finite axial detectors must be deemed impractical.

3.2 First moment detector. An alternative approach to phase contrast in a STEM is to employ multiple detectors, the final signal being a weighted difference signal from the individual channels. Such a detector may be constructed with a narrow $g(r)$, and one such system which accomplishes this ideally has already been proposed [8]. If a detector can be constructed with a response function $R(k) = \text{constant}$, $g(r)$ is infinitely narrow and has the form $g(r) = \text{constant} \times \delta(r)$. The vector nature of $R(k)$ simply implies two responses, one for each of the $x$ and $y$ directions. Although, as stated, the width of $g(r)$ is zero, phase contrast still results because of the differentiating effect of this function. In fact, the image signal can be found from eq. (3) without the need for any restrictions on $a$ or $\varphi$ to be [8]

$$s(r_0) = (2\pi)^{-1} [a^2 \varphi + b^2 - b^2 \varphi \gamma + a^2]$$

where we have taken $\gamma_0(r) = b(r) \exp(i \gamma(r))$.

Two things should be noticed about such a system. Firstly, it is bilinear in the quantities $a^2 \varphi$ and $a^2$, rather than the more usually considered quantities, $\varphi$ and $(1 - a)$, and secondly it is neither useful nor convenient to

![Fig. 2. Variation of computed Fresnel image intensity as a function of the semi-angle subtended by the detector, $\beta$; $x = 6 \times 10^7$ radian m$^{-1}$, $y = 5 \times 10^{-11}$m$^4$.](image-url)
classify it into a bright or dark field imaging mode. This classification is generally of little use when strong modulation exists.

3.3 Split detector. Whilst the first moment detector system provides an ideal method for imaging strongly modulating objects, it is instructive to study the behaviour of a more easily realisable and closely related system. This is the split-detector system introduced by Dekkers and de Lang [9, 10] and further discussed by Rose [11] and Hawkes [12]. The linear transfer properties of both this and the first moment system are discussed in the next section, but here we consider the ability of the split-detector to resist non-linearity in the formation of images of strong phase objects.

An infinite split-detector has a detector response function given by

\[ R(k) = \text{sgn}(k_x) \]

(9)

where

\[ \text{sgn}(k_x) = -1, \; k_x < 0 \]

\[ = 0, \; k_x = 0 \]

\[ = 1, \; k_x > 0 \]

Thus the corresponding function \( g(r) \) is given by

\[ g(r) = -i \left(2\pi x\right)^{-1} \delta(y) \]

(10)

Although this has no clearly defined width in the \( x \) direction it would still be expected that the major contribution to the integral over \( r' \) in eq. (5) would come from around \( g(0) \).

To study this further we again consider the simple phase function shown in fig. 1. In section 3.1 \( \Psi_0(k) \) was taken as \( \exp\left( iyk^2\right) \) and \( S(k) \to \delta(k) \) as \( \gamma \to 0 \) indicating that there is no contrast for zero defocus. With \( R(k) \) as in eq. (9) and \( \gamma \to 0 \) it is easy to show that

\[ S(k_x) = i\pi(\pi^2k_x)^{-1} \]

(11)

We observe that increasing \( \chi \) by a constant factor \( c \) merely increases \( S(k_x) \) by the same amount, and furthermore, as the object chosen was defined by \( dp/dx = \pi \text{sgn}(x) \), that \( S(k_x) \) is directly proportional to \( \mathcal{F}(dp/dx, x \to k_x) \). Thus an idealised split-detector gives a signal which is a faithful representation of the phase gradient of the transmittance, when this is as shown in fig. 1, irrespective of the magnitude of \( \chi \).

To investigate the effects of aberrations and apertures in the probe forming system, recourse was once again made to computer synthesis techniques. In contrast to the Fresnel case, even when appreciable non-linearity was present, differences in images obtained when the sign of \( \chi \) was changed were restricted to a reversal of contrast. That significant non-linearity was present in some cases, however, was determined by multiplying the phase function by \( c \) and testing whether or not the image signal scaled similarly. Some results of this are shown in fig. 3a, b where the phase functions and image
signals have been scaled and descaled respectively by 1, 2 and 4. Complete linearity would imply identity of the three descaled images and whilst this is clearly not the case, differences are not particularly pronounced. It is worth noting that the strongest phase object considered corresponds to a value of $\pi$ greater than those normally experienced experimentally and that the weakest of the phase structures corresponds to the value used in fig. 2 where non-linearity is clearly apparent.

Figure 3c shows images calculated in the same way as before but with the phase gradient step function replaced by a smoothly varying function which

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**Fig. 3.** Computed split-detector image of (a) test object of fig. 1 with $C_i = 0$, $Z = 0$ and semi-angle of the incident probe, $\alpha = 5 \times 10^{-4}$ radian; (b) as (a) but with $C_\theta = 100 \text{ m}\mu$; (c) a test object defined by $\frac{d\phi}{dx} = \pi \tanh (x/a)$ with $'a' = 2 \text{ nm}$, but otherwise with conditions as defined for (b).

The image profiles were normalised by dividing by 1, 2 and 4 (as indicated on the figure) corresponding to values of $\pi$ of $6 \times 10^9 \text{ radian m}^{-1}$, $1.2 \times 10^9 \text{ radian m}^{-1}$ and $2.4 \times 10^9 \text{ radian m}^{-1}$ respectively.
is a closer approximation to the spatial variation of induction in a domain wall. In fact, we chose \( dq/dx = \xi \tanh (x/a) \) and a value of \( 'a' \) of 2 nm, which is small for all but walls in the hardest magnetic materials. Comparison of the three parts of fig. 3 shows that non-linear effects are comparable in the unaberrated and very significantly aberrated cases chosen, indicating that for this detector geometry, the resistance to non-linearity depends on the function \( \rho(r) \) rather than \( \varphi(r) \).

So far we have only considered the problem of imaging one particular phase object, namely a more or less abrupt discontinuity in phase gradient. A rather different example is to assume an object, the phase of whose transmittance is given by

\[
q(x) = A \sin (2\pi vx)
\]

and here we calculate the response of a one dimensional split-detector system as a function of both \( A \) and \( v \). Neglecting aberrations as in previous examples, but assuming an aperture limited system so that

\[
\mathcal{F}_0(k_z) = 1, \quad |k_z| < k_0
\]

\[
= 0, \quad |k_z| > k_0
\]

it may be shown that, for integers satisfying \( 0 < p < 2k_0/v \), eq. (6) yields

\[
S_{(pv)} = \sum_m J_{m+p}(A) J_m(A) (2m + p) v, \quad -k_0/v < m < k_0/v - p
\]

\[
= -\sum_m J_{m+p}(A) J_m(A) (2k_0 - pv), \quad m < -k_0/v
\]

\[
= \sum_m J_{m+p}(A) J_m(A) (2k_0 - pv), \quad m > k_0/v - p.
\]

Here \( J_m \) is the \( m^{th} \) order Bessel function, and because of the periodicity of the transmittance, the spectrum \( S(k_z) \) is discrete. In fact, eq. (13) may very readily be generalised to a form appropriate to any object whose transmittance is periodic, satisfying \( h(x) = h(x + 1/v) \). If \( H(k_z) = \mathcal{F}(h(x), x \rightarrow k_z) = \sum_n \alpha_n \delta(k - n) \) the appropriate form of eq. (13) is obtained by replacing \( J_{m+p}(A) \) and \( J_m(A) \) by \( \alpha_{m+p} \) and \( \alpha_m \) respectively.

For the phase sinusoid of interest at present, we would expect \( S_{(pv)} \) to be zero for \( |p| = 1, \) if the imaging system is linear. The extent to which this is true has been evaluated numerically as a function of \( A \) and \( v/2k_0 \), and described elsewhere [8, 13]. Here we examine the linearity criterion of concern throughout this section, namely, if \( A \rightarrow cA \) does \( S_{(pv)} \rightarrow cS_{(pv)} ? \). By restricting attention to \( |p| = 1, \) and after considerable manipulation involving recursion relations between Bessel functions, eq. (13) reduces to the relatively simple form

\[
S_{(v)} = v(2k_0/v - 1) A [J^2_0(A) + J^2_1(A)]
\]

\[
-2v \sum_{m=1}^{M} (2k_0/v - 2m) J_{m-1}(A) J_m(A), \quad k_0/v > 1
\]

\[
= v(2k_0/v - 1) A [J^2_0(A) + J^2_1(A)], \quad 1/2 < k_0/v < 1
\]
where $M$ is the integral part of $k_0/\pi$. This may easily be evaluated as a function of $A$ and $\nu/2k_0$, and fig. 4 shows how the amplitude of the phase sinusoid at which $S(\nu)/A$ differs from its value for $A \ll 1$ by 5%, varies as a function of reduced spatial frequency. Thus, even for high spatial frequencies the system responds approximately linearly for phase excursions $\sim 1$ radian, and for lower spatial frequencies its performance is even better.

4. Linear transfer characteristics of differential phase contrast systems

As we have seen in the previous section, the first moment and split-detector systems are closely related, both giving rise to differential phase contrast. This similarity can best be demonstrated by studying their transfer functions and throughout this section linear imaging conditions are assumed. It is easily seen from eq. (8) that the point spread function for the quantity $\sigma^2 \nu \varphi$ in the first moment system is the same as the amplitude point spread function for a STEM with a large uniform detector. The principle of equivalence therefore ensures that the corresponding transfer function is the same as the incoherent spatial transfer function given by Black and Linfoot [14].

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Fig. 4. Contour of the maximum acceptable amplitude of phase sinusoid ($A_{\text{max}}$) as a function of reduced spatial frequency ($\nu/2k_0$). By maximum acceptable is meant the value at which $S(\nu)/A$ differs by 5% of the value for $A \ll 1$. 

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It is generally more convenient, however, to compare the two systems if we consider only pure phase objects as this allows the transfer function for phase (rather than \( a^2 \) \(|g|\)) to be displayed. The phase-contrast transfer function for the split-detector system may be calculated by assuming a weak sinusoidal phase grating as an object. Equation (3), after some simple manipulation, can then be used to give the phase-contrast transfer function as

\[
T_T(k) = i \iiint R(k') \langle \Psi_0(k') \Psi_0^*(k' - k) - \Psi_0(k') \Psi_0^*(k' + k) \rangle \, dk'
\]  

(15)

Fig. 5. Phase-contrast transfer functions for (a, c, e) a STEM with a split-detector; 
(b, d, f) a STEM with a first moment detector: 
\( A = 2 \); for (a) and (b) \( B = -1 \); for (c) and (d), \( B = -0.5 \); for (e) and (f) \( B = 0 \).
Some examples of the on-axis values of the function (namely $T_{\psi d}(k_x, 0)$) have been given by Dekkers and de Lang [10] whilst Rose [11] gives the two-dimensional function for the unaberrated case. Figures 5a, c, e show some phase-contrast transfer functions for an aberrated STEM using a split-detector and figs. 5b, d, f show the corresponding cases for a first moment system. All these transfer functions are purely imaginary and antisymmetric with respect to $k_x$, corresponding to differentiation in this direction with some spatial filtering. The following parameters are used

$$A = \frac{C_s \alpha^4}{4 \lambda}, \quad B = \frac{2Z}{C_s \alpha}, \quad 2k_0 = \frac{2\pi}{\lambda} \quad (16)$$

where $C_s$ is the spherical aberration coefficient and $\alpha$ the illumination semi-angle. For comparison fig. 6a and 6b show the transfer functions for an unaberrated case.

The phase-contrast transfer functions are, as expected, very similar for the two detector geometries, and both have the advantage that no unwanted bands of contrast reversal occurs. Figures 5c and 5d show that the spherical aberration can be partially balanced with defocus, although this results in lower resolution. Small but significant differences do exist, however, between the phase-contrast transfer functions of the two systems. The most important of these are: (i) the first moment system weights lower spatial frequencies (at least on the $k_x$ axis) more highly than the split-detector. (ii) The transfer function for $dq/dz$ is rotationally symmetric for the first moment detector but only centrosymmetric in the split-detector. The amplitude-contrast transfer functions are also similar. These can be calculated by considering a weak sinusoidal amplitude grating which results only in a simple change of eq. (15). Both systems would give zero amplitude contrast if an unaberrated probe was used and for both systems amplitude contrast increases with aberrations. Figures 7a and 7b show the amplitude-contrast transfer functions for the two systems when the balanced condition applies, as in figs. 5c and 5d. A significant difference between the two amplitude-contrast transfer functions is that the split-detector system has spatial frequency cut off on the $k_x$ axis at

![Fig. 6. Phase-contrast transfer functions for (a) a STEM with a split-detector and (b) a STEM with a first moment detector. The probe forming system is assumed unaberrated.](image)
half that for the phase-contrast transfer function whilst the first moment system has not.

5. Conclusions

It has been demonstrated that the linear imaging of strong phase objects depends not only on the nature of the phase function, but also on the electron optical system used. In particular, by employing a STEM with an anti-symmetrical detector, an equivalent degree of coherence may be produced which is both oscillatory and localised. This results in the required ability to linearly image strong phase objects, and hence greatly simplifies the interpretation of images of such objects. The transfer characteristics of an ideal system of this type (the first moment detector system) are shown to be closely related to those of the split-detector system of Dekkers and de Lang. Both of these systems have transfer functions with desirable properties.

Acknowledgements

We are very grateful to Professor R.P. Ferrier for encouragement and advice throughout this work.

Fig. 7. Amplitude-contrast transfer functions for (a) a STEM with a split-detector and (b) a STEM with a first moment detector. All parameters as in fig. 5c, d.

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