Quantitative measurements of phase using the transport of intensity equation

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Abstract. In this paper we present considerations for experimental recovery of electron phase using the TIE algorithm as applied to TEM images. Realistic simulations have considerably aided interpretation of results. Experimental reconstructions show that low spatial frequency artefacts in the phase reconstruction, resulting from very small variations in the detected signal, can be reduced by using images with smaller numbers of pixels.

1. Introduction
The transport of intensity equation (TIE) applied to transmission electron microscopy (TEM) allows, in principle, reconstruction of the phase of the electron beam [1,2]. This technique is classed as a non-interferometric method and can therefore provide a simpler route to phase information than electron holography. Since the method has been proposed over 20 years ago a number of materials systems have been successfully studied [3,4,5,6]. However the method has not become as widely used as might have been expected due to a number of problems. One obstacle is that the range of applicability is limited to the range of linear imaging. In general for high resolution, i.e. transferring high spatial frequency components linearly, the defocus values required are rather low. Consequently the phase contrast is low and therefore the signal to noise ratio of phase information in the defocused image is a serious issue.

Additionally there are many practical issues which need to be treated very carefully in order to confidently obtain accurate phase information. In principle the method requires 3 images at equally spaced defocus and therefore accurate image alignment and registration is a major concern. Single pixel misalignment will result in artefacts in the reconstructed phase. Furthermore the TIE reconstruction algorithm is effectively a low spatial frequency filter and artefacts at lower frequencies due to very small intensity variations in the images can dominate the reconstructed phase [4,5].

In this paper we consider a simple electrostatic phase sample and investigate some of the issues for successful phase reconstruction. Image simulation can easily be carried out for such a system. Comparison with experimental images confirm the degree of success of the TIE method. Furthermore it is shown that low spatial frequency artifacts can be reduced by using a smaller image size.

2. Image calculations and comparison with experiment
The system to which we apply the TIE method here is a standard holey carbon sample (from Agar Scientific). Assuming the carbon film is of constant thickness the phase can then be represented by a top-hat function with a finite slope at the edges [4]. For images calculated here the pixel spacing is
taken as 2.5 nm and the variation over the edge is assumed linear and occurs over 4 pixels (10 nm). Calculation of the phase variation due to the mean inner potential, V, of the film uses the standard equation \( \phi = \pi (Vt/\lambda E) \) where \( t \) is the sample thickness, \( \lambda \) the electron wavelength and \( E \) the accelerating voltage of the TEM. The images here are to be compared with those taken on a Philips CM20 TEM with \( E = 200 \text{kV} \) and so has \( \lambda = 2.5 \text{ pm} \). Carbon has \( V = 9.09 \text{V} \) and for this sample the thickness is \( \sim 20 \text{ nm} \) (confirmed by electron energy loss spectroscopy measurements). This therefore gives a phase jump of 1.14 radians (just over \( \pi/3 \) radians) between the film and vacuum.

Calculation of the simulated images was made using scripted programs we have written which run on the imaging and analysis software package Digital Micrograph™ from Gatan. The parameters used for imaging here are equivalent to the TEM conditions in the CM20 for Lorentz mode i.e. with the objective lens switched off and the sample in field free space. There are a number of reasons for using this mode, firstly it is intended to show TIE works reasonably for medium resolution here and secondly we intend using the TIE mainly for magnetic phase reconstructions in future. The conditions input to the image calculation script are defocus, pixel spacing, coefficient of spherical aberration (8000 mm for the Lorentz lens) and beam convergence angle (10 \( \mu \text{rad} \) is used here). An example of a calculation of simulated Fresnel image, using a suitable function also to represent the amplitude is shown in Fig. 1(a) for a defocus of \( +160 \\mu \text{m} \). An equivalent experimental image taken in the CM20 microscope is shown for comparison in Fig. 1(b).

![Fig. 1 (a) Simulated and (b) experimental Fresnel images of a holey carbon film taken at defocus values of 160 \( \mu \text{m} \) in field free mode in the TEM.](image)

3. TIE phase reconstruction for holey carbon film

The simulated and experimental images are then used with the TIE algorithm to calculate the phase of the electron beam. In the case of the experimental images great care must be taken to ensure any magnification change due to defocus is corrected in addition to scaling for changes in intensity. We now consider the range of application as it has been noted that the TIE reconstruction appears to be robust beyond the strict definition of defocus condition [4,6]. Strictly speaking the TIE applies for linear Fresnel imaging which suggests the condition is \( \pi \lambda f k^2 << \pi \) where \( f \) is the defocus distance and \( k \) the highest spatial frequency of interest. As this is a manifestation of the small angle formula for trigonometric functions the equation may be set equal to \( \pi/10 \) and we can then calculate the maximum defocus allowed for a given spatial frequency. Note that some authors set a less stringent condition for this equation [4,5]. So the definition we have derived for linear imaging is \( f_{L1} = 1/(10\lambda k^2) \) whilst for the other definition it is \( f_{L2} = 1/(\pi \lambda k^2) \).

The spatial frequency components present in the images of the holey carbon film need to be considered. The highest spatial possible frequency for the pixel spacing corresponds to \( k_{max} = (5\text{nm})^{-1} \). However this results in a very restricted range of values of \( f_{L1} = 1 \mu\text{m} \) and \( f_{L2} = 3.2 \mu\text{m} \). These small
focus values would be required if an accurate phase profile on the pixel scale is required. In the situation here we can define further the spatial extent of the phase variation of the edge of the hole (10 nm) which gives us defocus values $f_{L1} = 4 \mu m$ and $f_{L2} = 12.7 \mu m$. The limit of defocus is open to interpretation and is quite dependent of the spatial frequency spectrum of the phase object. For the holey carbon film considered here one can consider using TIE to recover accurately the phase profile in which case the focus values used for the spatial extent of the edge should be used. However if one only wants to measure the phase change between the film and vacuum then much larger focus values may be used as shall be shown later in this paper.

For the situation here we have also found that we encounter unwanted low spatial frequency signal variation in the recovered TIE phase from the experimental images consistent with previous studies [4,6]. The source of these variations is very small intensity changes in the images, which may be due to the gun, sample or detector. However this effect is considerably reduced when using images at larger defocus values. A small intensity gradient will give a very large low spatial frequency contribution using TIE at small defocus, but the same gradient present in a larger focus image will have a much smaller contribution to the TIE reconstruction. We are working on quantifying this effect.

In Fig. 2 the recovered phase is shown for simulated and experimental images taken with defocus values of 160 µm. A comparison of the input simulated phase (phase jump over 10 nm) and the TIE phase at 160 µm indicates that the edge profile has been broadened as expected but that the phase jump between inside and outside of the whole has been well preserved. In this case the limit of spatial frequency for the two focus limits ($f_{L1}$ and $f_{L2}$) are $(63\text{nm})^{-1}$ and $(35\text{nm})^{-1}$. This confirms the “robustness” of the TIE method as long as one is aware of the spatial frequency limitation. There is still some spurious low spatial frequency contribution in the experimental image though it is much less than that observed at smaller defocus values.

At lower defocus values the low contrast and contributions from “noise” in the images causes more serious problems. To highlight this effect we consider the phase reconstructions from a hole using the same images but in one case using an area of 512×512 centered on the hole whilst in the other an area of 256×256 pixels is used, the same pixel spacing is used in each case. These areas were defined from original images of 1024×1024 pixels. Intuitively one would expect the phase variation in the reconstructions should be the same, however in experimental images the noise spectra means that the low spatial frequency noise or artefact contribution in the larger area images is considerably greater. The TIE reconstructions from these areas are shown in Fig. 3 from a defocus set of images at ±80 µm. From the smaller area (Figs 3(a) and (b)) it can be seen that although the edge profile is again broadened the phase profile resembles a top hat function. The phase jump is well reproduced at $-\pi/3$. From the same hole but using a larger surrounding area (Figs 3(c) and (d)) low spatial frequency artefacts are clearly visible. Although the phase jump can still be discerned the magnitude of the artefact is larger than the phase jump itself. Evidently the increased contribution from low spatial

![Fig. 2](image-url) (a) Simulated and (b) experimental TIE recovered phase from Fresnel images of holey carbon film taken at defocus values of 160 µm.

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frequency “noise” has a considerable detrimental effect on the reconstruction. Again this is an area that requires further detailed study.

4. Conclusion

We have shown that the TIE method works best for medium resolution, i.e. larger defocus values, if the low spatial frequency signals are to be suppressed. Additionally low spatial frequency noise artefacts can be reduced by using smaller images sizes. Although reducing these effects is possible, it is very challenging measuring small phase changes with the TIE method. In future we intend to use the TIE method principally for magnetic phase reconstruction where the phase gradient provides information on the magnetic induction distribution. As the differential operator provides suppression of the low spatial frequency signal it is expected that the TIE method should be well suited to such systems.

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