X-ray phase imaging: Demonstration of extended conditions with homogeneous objects

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Abstract: We discuss contrast formation in a propagating x-ray beam. We consider the validity conditions for linear relations based on the transport-of-intensity equation (TIE) and on contrast transfer functions (CTFs). From a single diffracted image, we recover the thickness of a homogeneous object which has substantial absorption and a phase-shift of -0.37 radian.

OCIS codes: (340.7480) X-rays; (340.7440) X-ray imaging; (340.7460) X-ray microscopy

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1. Introduction

Absorption contrast has been the principal imaging mode for x-rays for over 100 years, nonetheless there has been considerable development recently in implementing phase contrast techniques. Phase contrast can be strong when absorption contrast is minimal; for instance for low-Z materials or for high energy x-rays [1, 2, 3]. Phase contrast can also be used without delivering a high dose to the sample [2]. And certain phase methods require no additional optics leading to source-limited, rather than optics-limited, resolution [4].

Demonstrated methods for obtaining the phase from an x-ray wavefield are now legion. Interferometric methods include the use of Bonse and Hart [5] type interferometers [6], shearing interferometers [7], differential interference contrast arrangements using two zone plates [8] or diffractive optical elements [9]. Zernike phase contrast [1] has also been demonstrated. Other methods include refraction measurement using crystal diffraction [10], and segmented detectors [11]. Propagation-based methods have also been developed and involve recovery of the object phase and/or amplitude from one or more measurements of the object diffraction pattern. Methods include in-line holography [12], iterative schemes [13] and approaches based on solution of the equations governing the propagation of the wavefield [14, 15, 3].

In this paper we consider aspects of the latter method. Here the Fresnel integral describing the diffracted intensity is linearized allowing a straightforward retrieval of object phase and transmission. A first order Taylor expansion obtains the transport of intensity equation (TIE) solution [16, 3]. Alternatively a Born-type approximation [17, 16] gives a solution identical in form to that derived by Guigay [18, 19] in the context of electron microscopy. We will refer to this as the contrast transfer function (CTF) solution. Both the TIE and the CTF solution can be further simplified under the assumption of a homogeneous object [20, 16]. This permits the thickness distribution of an object to be retrieved from a *single* diffracted image.

In the TIE it is the first-order Taylor expansion that restricts the validity of the solution. Other than a requirement for paraxiality, there is no limitation on the magnitude of the phase or the absorption. On the other hand, the Born-type approximation previously used in deriving the CTF method can be quite restrictive. Guigay showed [19] that a less restrictive requirement applies for a pure phase object. Like the TIE condition, this condition depends on the feature sizes present as well as the wavelength and propagation distance.

Here we show that the Fresnel-diffracted intensity can be linearized at a later stage of the derivation. As a consequence we find that the less restrictive phase condition extends to weakly-absorbing objects. In Section 2 we outline the derivations of the TIE and the CTF solutions and their validity conditions and show how the assumption of a homogeneous object allows retrieval of the object thickness from a single diffracted image. In Section 3 we present experimental results demonstrating quantitative thickness retrieval under our derived validity conditions.

2. Derivations

2.1. TIE and CTF

We begin with the Fourier transform, $\mathscr{F}[\cdot]$, of the intensity $I(\mathbf{r},z)$ obtained under Fresnel diffraction of an object-plane wavefield $f(\mathbf{r}) = f(\mathbf{r},z=0)$ with wavelength λ [18]:

$$\mathscr{F}[I(\mathbf{r},z)] = \int_{-\infty}^{+\infty} f^*(\mathbf{r}+\lambda z\mathbf{u}/2)\, f(\mathbf{r}-\lambda z\mathbf{u}/2)\exp(-2\pi i\mathbf{r}\cdot\mathbf{u})\,\mathrm{d}\mathbf{r}. \tag{1}$$

The transverse spatial coordinates and their corresponding Fourier conjugates are given by $\mathbf{r}$ and $\mathbf{u}$ respectively. To obtain the TIE solution we Taylor expand the wavefield to first order,

$$f(\mathbf{r}+\lambda z\mathbf{u}/2) = f(\mathbf{r}) + \frac{1}{2}\lambda z\mathbf{u}\cdot\nabla f(\mathbf{r}), \tag{2}$$

where ∇ is the gradient operator in the transverse plane. Substituting into Eq. (1) gives

$$\nabla\cdot(I(\mathbf{r},z)\nabla\phi(\mathbf{r},z)) = -\frac{2\pi}{\lambda}\frac{\partial}{\partial z}I(\mathbf{r},z), \tag{3}$$

where ϕ is the phase of the wavefunction f. The validity condition for the TIE solution is therefore that the higher order Taylor expansion terms can be disregarded:

$$\left|\sum_{j=2}^{\infty}\frac{1}{j!}\left(\frac{1}{2}\lambda z\mathbf{u}\cdot\nabla\right)^j f(\mathbf{r})\right| \ll 1. \tag{4}$$

This condition can always be satisfied by choosing a sufficiently small propagation distance, while requiring no approximation regarding the magnitude of the amplitude or phase.

The previous approach to obtain the CTF solution was to write the object wavefunction in terms of its absorption μ and phase ϕ components such that:

$$f(\mathbf{r}) = f_0\exp(-\mu(\mathbf{r}) + i\phi(\mathbf{r})), \tag{5}$$

where $I_0 = |f_0|^2$ is the intensity of the plane wavefield incident on the object. Then the Born-type approximation of $\mu \ll 1$ and $|\phi| \ll 1$ was made so that

$$f(\mathbf{r}) = f_0(1-\mu(\mathbf{r}) + i\phi(\mathbf{r})). \tag{6}$$

Substituting Eq. (6) into Eq. (1) and retaining μ and ϕ to first order, obtained Guigay's result

$$\mathscr{F}[I(\mathbf{r},z)] = I_0\left(\delta(\mathbf{u}) - 2\cos(\pi\lambda z u^2)\mathscr{F}[\mu(\mathbf{r})] + 2\sin(\pi\lambda z u^2)\mathscr{F}[\phi(\mathbf{r})]\right) \tag{7}$$

in which $\delta(\mathbf{u})$ denotes the Dirac delta distribution. When the object is pure phase ($\mu = 0$), Guigay [19] also showed, by substituting Eq. (5) into Eq. (1), that the corresponding form of Eq. (7) can be obtained if, for all $\mathbf{r}$,

$$|\phi(\mathbf{r}+\lambda z\mathbf{u}/2) - \phi(\mathbf{r}-\lambda z\mathbf{u}/2)| \ll 1. \tag{8}$$

This is sometimes referred to as the slowly-varying phase condition. It should be noted that the displacement vector $\lambda z\mathbf{u}$, over which points in the phase should be similar, is a function of propagation distance z and of spatial frequency $\mathbf{u}$.

2.2. TIE for a homogeneous object

Consider the case of an optically thin and homogeneous object for which

$$\mu(\mathbf{r}) = k\beta T(\mathbf{r}) \quad \text{and} \quad \phi(\mathbf{r}) = -k\delta T(\mathbf{r}), \tag{9}$$

where $k = 2\pi/\lambda$, the refractive index is $n = 1-\delta+i\beta$ and T is the thickness of the object. We substitute into Eq. (1), re-factor and make a first-order Taylor expansion of T yielding

$$\mathscr{F}[I(\mathbf{r},z)] = I_0 \int_{-\infty}^{+\infty} \exp(-2k\beta T(\mathbf{r}))\exp(-ik\delta\lambda z\mathbf{u}\cdot\nabla T(\mathbf{r}))\exp(-2\pi i\mathbf{r}\cdot\mathbf{u})\,\mathrm{d}\mathbf{r}. \tag{10}$$

If we assume that $|\lambda zu\cdot\nabla\phi(r)| \ll 1$ then we can expand the second exponential to first order,

$$\mathscr{F}[I(\mathbf{r},z)] = I_0 \int_{-\infty}^{+\infty} \exp(-2k\beta T(\mathbf{r}))\,(1-ik\delta\lambda z\mathbf{u}\cdot\nabla T(\mathbf{r}))\exp(-2\pi i\mathbf{r}\cdot\mathbf{u})\,\mathrm{d}\mathbf{r} \tag{11}$$

and then applying the Fourier derivative theorem $\mathscr{F}[\nabla f(\mathbf{r})] = 2\pi i\mathbf{u}\mathscr{F}[f(\mathbf{r})]$ we obtain:

$$\mathscr{F}[I(\mathbf{r},z)] = I_0\mathscr{F}[\exp(-2k\beta T(\mathbf{r}))]\left(1+\frac{\delta}{\beta}\lambda zu^2\right). \tag{12}$$

This may be solved for the thickness T:

$$T(\mathbf{r}) = -\frac{1}{2k\beta}\ln\mathscr{F}^{-1}\left[\frac{\beta}{\beta+\delta\pi\lambda zu^2}\mathscr{F}\left[\frac{I(\mathbf{r},z)}{I_0}\right]\right]. \tag{13}$$

The validity condition on the thickness T is similar to that for the TIE in Eq. (4).

2.3. Derivation of extended validity CTF solution

Here we begin by substituting Eq. (5) into Eq. (1):

$$\mathscr{F}[I(\mathbf{r},z)] = I_0 \int_{-\infty}^{+\infty} \exp\left(-\mu(\mathbf{r}+\lambda z\mathbf{u}/2)-\mu(\mathbf{r}-\lambda z\mathbf{u}/2)+i\left(\phi(\mathbf{r}-\lambda z\mathbf{u}/2)-\phi(\mathbf{r}+\lambda z\mathbf{u}/2)\right)\right) \times \exp(-2\pi i\mathbf{r}\cdot\mathbf{u})\,\mathrm{d}\mathbf{r}. \tag{14}$$

Assuming both real and imaginary parts of the exponential are small, we expand, noting the Fourier transforms:

$$\begin{aligned}\mathscr{F}[I(\mathbf{r},z)]/I_0 = \delta(\mathbf{u}) - &\mathscr{F}[\mu(\mathbf{r}+\lambda z\mathbf{u}/2)+\mu(\mathbf{r}-\lambda z\mathbf{u}/2)] \\ &+ i\mathscr{F}[\phi(\mathbf{r}-\lambda z\mathbf{u}/2)-\phi(\mathbf{r}+\lambda z\mathbf{u}/2)].\end{aligned} \tag{15}$$

Applying the Fourier shift theorem $\mathscr{F}[f(\mathbf{r}-\mathbf{a})] = \exp(-2\pi i\mathbf{a}\cdot\mathbf{u})\mathscr{F}[f(\mathbf{r})]$ to each term and rearranging recovers Eq. (7). If the object is homogeneous then, substituting Eq. (9), we can retrieve the thickness T from a single diffracted image:

$$T(\mathbf{r}) = \mathscr{F}^{-1}\left[\frac{1}{-2k\left(\delta\sin(\pi\lambda zu^2)+\beta\cos(\pi\lambda zu^2)\right)}\mathscr{F}\left[\frac{I(\mathbf{r},z)}{I_0}-1\right]\right]. \tag{16}$$

The linearizing assumption made in obtaining Eq. (15) is that

$$2\mu(\mathbf{r}) \ll 1 \quad \text{and} \quad |\phi(\mathbf{r}+\lambda z\mathbf{u}/2)-\phi(\mathbf{r}-\lambda z\mathbf{u}/2)| \ll 1. \tag{17}$$

These conditions on the validity of Eq. (7) are much less stringent than previously realised. While the object must be weakly absorbing it need not be non-absorbing, which is less restrictive than the pure phase assumption made by Guigay. Furthermore, the weak phase condition required in the Born-type approach is here relaxed to the slowly-varying condition Eq. (8).

Assuming object homogeneity allows us to incorporate the effect of object absorption. Consequently the contrast transfer function, which is the denominator term of Eq. (16), is non-zero at $u = 0$. Nulls in the CTF at frequencies above $u = 1/\sqrt{2\lambda z}$ may be avoided by incorporating additional images at different propagation distances [21, 22], or simply by Tikhonov regularisation [23]. We find that Tikhonov regularisation leaves only minor artefacts at high frequencies whereas assuming a pure phase object introduces intractable low-frequency instability.

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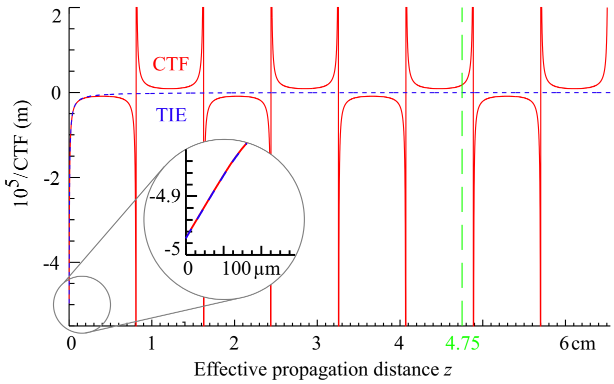

Fig. 1. Inverse of the contrast transfer functions for the TIE (blue dotted line) and CTF (red line) forms calculated for an infinite grid with spatial feature size of 1.9 µm and the experimental conditions. The green dashed line is at the experimental distance. The embedded movie shows the inverse CTFs as shown here with a slider indicating the z position corresponding to diffraction distance. Also shown in the movie is a plot of an input amplitude (green) and the amplitude retrieved using either the CTF (red) or TIE (blue) methods for the indicated propagation distance. The CTF method correctly accounts for the contrast reversals that arise on propagation. The TIE method should only be applied for z closer than the first contrast reversal; it may retrieve *inverted* amplitudes if applied at greater z.

3. Experimental results

Laser ablation [24] was used to etch a grid of lines in a polyimide film (composition $C_{22}H_{10}N_2O_4$ and density 1.45 g cm^{-3}). The lines were measured by atomic force microscope (AFM) to be approximately 1.9 µm apart and 90 nm high and were superimposed on an 80 µm square of 650 nm in height.

The experiments were performed at a wavelength of 0.436 nm and with $z = 0.0475$ m, where z is as defined above. Through the thickest part of the object the transmission $\exp(-2\mu)$ is 98.7% and the phase-shift ϕ is -0.37 radian. Accordingly, neither the Born-type approximation nor the phase-only requirement are met. However, for most positions and spatial frequencies the slowly-varying phase condition Eq. (8) is obeyed.

Figure 1 shows a plot of the inverse of the contrast transfer function from the CTF solution Eq. (16) for the 1.9 µm grid features. A contrast transfer function can also be defined, in the weak absorption limit, for the TIE solution Eq. (13) and its inverse is also shown in Fig. 1. Inspection shows that the TIE solution should yield grid features reversed in contrast compared to the CTF solution. No contrast reversal is expected for the 80 µm square.

The experiments were performed at beamline 2-ID-B at the Advanced Photon Source. A beam of 2844 eV ($\Delta E/E \simeq 10^{-3}$) x-rays with FWHM size 1.5×0.5 mm illuminated a 160 µm diameter gold zone plate with an outer zone width of 50 nm. The focal length was 18.4 mm and a 10 µm diameter order sorting aperture was placed at a distance of 17.2 mm from the zone plate. A 30 µm wide gold beamstop blocked the zero order beam. This zone plate configuration provided a point source of illumination at a distance of $R_1 = 54.7$ mm from the object. After passing through the object the beam was allowed to propagate through a He-filled flight tube a distance of $R_2 = 363$ mm onto a crystal scintillator which was imaged through a 20× objective by a CCD camera with 13.5 µm pixels. This expanding beam mode is analyzed using the parallel beam mode derivations described above using the conversion [4]

$$I_{R_1}(\mathbf{r}, R_2) = \frac{1}{M^2} I_\infty \left(\frac{\mathbf{r}}{M}, \frac{R_2}{M} \right), \tag{18}$$

where $M = (R_1 + R_2)/R_1$ is the magnification, $I_{R_1}(\mathbf{r}, R_2)$ is the expanding beam diffraction-

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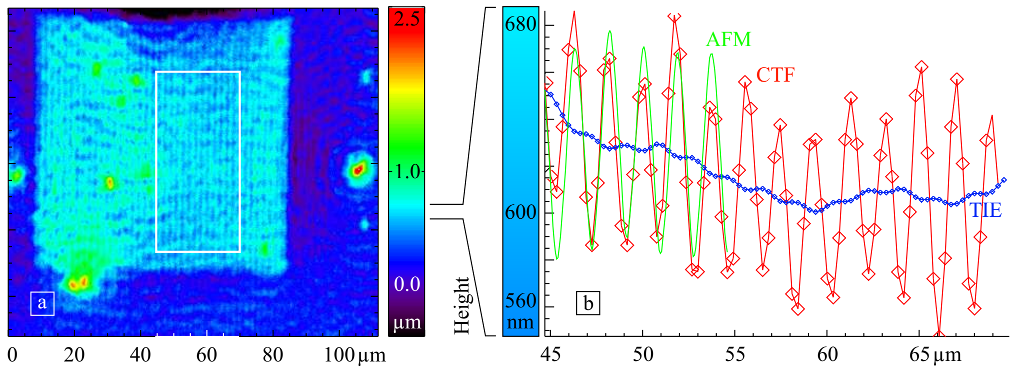

Fig. 2. (a) CTF-retrieved thickness map for the square with grid lines.(b) Column-average of retrieved thickness for the grid pattern in the region shown in (a) for the TIE solution (blue) and the CTF solution (red). The AFM result (green) shows excellent agreement. AFM measurements also confirm the presence of grid lines outside the square. These are not a retrieval artefact, unlike the circular fringes around the contaminant at centre right. The contaminating material presumably violates the assumption of an homogeneous object.

pattern due to a point source a distance R_1 behind the object and I_∞ is parallel beam diffraction-pattern. The object thickness map was retrieved from the measured diffraction pattern using both the CTF solution Eq. (16) with Tikhonov regularization, and the TIE solution Eq. (13), and is shown in Fig. 2(a). We line average the essentially one-dimensional image in the region outlined in white. Both CTF and TIE methods retrieve the mean height of the 80 μm square as 610 ± 50 nm, concurring with the AFM result of 650 nm. However, the TIE retrieval grossly underestimates the height of the grid pattern and, as predicted in Fig 1, reverses the contrast (Fig. 2(b)). As expected from the validity conditions for Eq. (16), the CTF retrieval of the grid pattern (Fig. 2(b)) is in excellent agreement with the AFM measurement of 90 nm.

4. Conclusions

We have explored the validity conditions for the linear CTF expression relating object phase-shift and absorption to the contrast of the Fresnel diffraction-pattern. The linear expression is found *not* to be restricted to weakly phase-shifting objects: it applies to a substantially wider class of objects which show weak absorption and slowly-varying phase.

If an object is made of one material with known complex refractive index, the CTF expression may be inverted to retrieve the object thickness from a single diffracted image. We demonstrated an example where the CTF solution could correctly retrieve thickness features of a weakly-absorbing object with large, but slowly-varying, phase-shift. Thickness features at two well-separated spatial frequencies were retrieved by the CTF solution while the TIE retrieval was valid only at the lower spatial frequency. These results augur well for wider applications of the CTF technique such as imaging cold atom clouds [25, 26] and plasmas [27].

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