Submicrometer Resolution Hard X-Ray Holography with the Asymmetric Bragg Diffraction Microscope

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The asymmetric Bragg diffraction microscope is a novel x-ray microscope which forms a magnified in-line near-field hologram by asymmetric reflection from two crossed flat crystals. In this paper, the optics of the microscope is studied theoretically. The optical transfer function is obtained, and the limiting spatial resolution, rated at 25% modulation transfer, is determined to be 0.30 μm at an optimum magnification of 89× with Si crystals, over a wide range of hard x-ray wavelengths. Absorption and phase contrast images can be computed from holograms acquired at several object distances. Application to submicrometer resolution hard x-ray microtomography is envisioned.

A quest to achieve 1 μm spatial resolution in hard x-ray (≥10 keV) microtomography using synchrotron radiation has spurred the development of detectors optimized for high resolution at the expense of detection efficiency. Currently, a state-of-the-art high resolution detector offers 0.8 μm resolution and 3% detection efficiency at 14 keV [1]. Even if a higher resolution detector could be developed, its efficiency might be too low to be generally useful, since low efficiency results in relatively noisy images or long exposures, and the latter may be impractical or produce excessive radiation damage in the object under study, particularly for biological materials.

A promising approach to achieve submicrometer resolution is to magnify the image with an x-ray optic and detect it with a low resolution, high efficiency detector, thus improving both resolution and efficiency. The optic must have submicrometer resolution, as well as adequate magnification, efficiency, and image fidelity. Only recently, two hard x-ray optics have demonstrated submicrometer resolution (0.3 μm): Fresnel zone plates [2,3] and parabolic compound refractive lenses [4]. However, they have not yet been used in hard x-ray microtomography.

One x-ray optic, the flat asymmetrically cut crystal [5], has been used in hard x-ray microtomography [6–9], but has not demonstrated submicrometer resolution. This optic originated in the field of x-ray diffraction, and its basic imaging properties, e.g., its limiting resolution, have been previously unknown. The purpose of this paper is to theoretically determine its imaging properties, and show they are conducive to performing submicrometer resolution hard x-ray microtomography with synchrotron radiation.

Bragg diffraction from an asymmetrically cut crystal produces one-dimensional magnification. In the simple case of coplanar diffraction [10] shown in Fig. 1, the magnification direction lies in the plane of diffraction [11] and the magnification factor is sin(θB + α) / sin(θB – α), where θB is the Bragg angle and α is the angle between the Bragg planes and surface. A second diffraction from another crystal with the same magnification factor but perpendicular plane of diffraction produces uniform two-dimensional magnification.

One-dimensional image magnification was first demonstrated by Kohra [12], who produced radiographic and topographic images at magnifications up to 5.5×, using CuKα radiation (8.0 keV). Compound one-dimensional magnification by double reflection in a grooved crystal was demonstrated by Bonse et al. [13], who obtained a resolution of 150 lp/mm (lp stands for line pair and represents a cycle of a spatial frequency) at 10% modulation transfer, using 8.2× (total) magnification and 10.3 keV synchrotron radiation. One-dimensional magnification was applied to computed tomography by Sakamoto et al. [6], Bonse et al. [7], and Nagata et al. [8], using synchrotron radiation.

Two-dimensional image magnification was first demonstrated by Boettinger et al. [14], who used two crystals with perpendicular planes of diffraction to produce 25× images with CuKα radiation. Another setup for two-dimensional magnification using only one crystal (but still two diffractions) was demonstrated by Korytar [15], who produced 6× images with CuKα radiation. For the latter setup, the problem of finding a pair of surfaces which produce images free of shear distortion was solved analytically by Dobrocka [16].

Using the above two-crystal setup, 79× magnification, and 12.3 keV synchrotron radiation, Kuriyama et al. [17] resolved a 420 lp/mm resolution target, implying a resolution of about 1.2 μm. They also observed Fresnel

FIG. 1. Asymmetric Bragg diffraction. Slanted lines below crystal’s surface represent Bragg planes.
diffraction effects in images, not reported previously, which were studied quantitatively by Spal [18]. Silver [9] performed computed tomography with this instrument using synchrotron radiation.

Only the two-crystal setup with perpendicular planes of diffraction is considered below, and the crystals, subsequently called magnifiers, are assumed to be identical. The incident beam is assumed to be a linearly polarized plane wave of wavelength $\lambda$ and wave number $k = 2\pi/\lambda$, whose electric field is parallel to the plane of diffraction of the first magnifier. Inelastically scattered and secondary radiation are ignored.

To analyze the microscope, rectangular coordinate systems are established at the object, magnifiers, and detector, as shown in Fig. 2. The positive $z$ axes of these systems follow the incident beam through the microscope, and pass through the centers of the object, magnifier surfaces, and detector. The magnifiers are aligned to pass this beam through the centers of their rocking curves. At each magnifier, two coordinate systems are established, denoted by CS$_{1h}$ for the $h$th magnifier, where $h$ is $O$ for the incident beam coordinate system and $H$ for the diffracted beam coordinate system. CS$_{1O}$ and CS$_{1H}$ share the same origin $O_1$ and $y$ axis, which lie on the magnifier’s surface, while CS$_{2O}$ and CS$_{2H}$ share the same origin $O_2$ and $x$ axis, also on the magnifier’s surface. The positive $x_{1O}$ and $y_{2O}$ axes are directed away from the magnifiers. The remaining coordinate systems are indicated by the subscripts $o$ and $d$, for object and detector. Coordinate systems with parallel $z$ axes (e.g., CS$_o$ and CS$_{1O}$) also have parallel $x$ and $y$ axes. The $y_o$ axis is assumed to be vertical.

The optical axis is the broken line joining all the origins $O_p$, and the distance from $O_p$ to $O_q$ measured along the optical axis is denoted $Z_{pq}$. The magnification factor of the $l$th magnifier, defined as $|\mathbf{n}_l \cdot \mathbf{z}_{2l}/\mathbf{n}_l \cdot \mathbf{z}_1|$ where $\mathbf{n}_l$ is a vector normal to its surface, is the same for both magnifiers and is denoted $m$. The vectors $\mathbf{r} = (x, y, z)$ and $\mathbf{s} = (x, y)$ are used to represent arbitrary points and points on the plane $z = 0$, respectively. The vectors $\mathbf{r'} = (x', y', z')$, where $z' = \sqrt{1 - x'^2 - y'^2}$, and $\mathbf{s'} = (x', y')$ are used to represent unit wave vectors and their transverse components, respectively.

It is convenient to decompose the electromagnetic field in each section of the microscope, defined by a segment of the optical axis, into an angular spectrum of plane waves with wave number $k$. Only paraxial plane waves, i.e., those with $|s'| \ll 1$, are considered, because others will be strongly attenuated by the magnifiers. Because the electric field polarization vector of each paraxial wave is very close to $\hat{x}$, any difference is ignored and the electromagnetic fields are treated as scalar fields. The electric field amplitude $E(\mathbf{r})$ and its angular spectrum $E'(s')$ are related by $E(\mathbf{r}) = \int ds' E'(s') \exp(ik\mathbf{r'} \cdot \mathbf{r})$, where the time dependence of $E(\mathbf{r})$ has been suppressed. $E(\mathbf{r})$ in the section between $O_p$ and $O_q$, where $CS_p$ and $CS_q$ are parallel, is denoted by $E_p(\mathbf{r}_p)$ when expressed in $CS_p$ coordinates, and by $E_q(\mathbf{r}_q)$ when expressed in $CS_q$ coordinates: e.g., $E_o(\mathbf{r}_o) = E_{1O}(\mathbf{r}_{1O})$ is the electric field amplitude between the object and first magnifier. The angular spectrum of $E_p(\mathbf{r}_p)$ is denoted by $E'_p(s'_p)$.

The structure of the object determines $E_o(\mathbf{r}_o)$ on the object plane, which in turn determines $E'_o(s'_o) = \lambda^{-2} \int ds_o E_o(s_o) \exp(-ik\mathbf{s}_o \cdot \mathbf{s}_o)$. Since $\mathbf{r}_p = \mathbf{r}_{1O} + Z_{1O}\mathbf{z}_o$, $E'_o(\mathbf{s}_{1O}) = \exp(ikZ_{1O}\mathbf{s}_o)E'_o(s'_o)$, where $s'_{1O} = s'_o$. The phase factor $\exp(ikZ_{1O}\mathbf{s}_o)$ represents Fresnel diffraction between the object and first magnifier.

At the first magnifier, according to the dynamical theory of x-ray diffraction [19], each component incident wave creates a component diffracted wave, producing $E_{1H}(\mathbf{r}_{1H}) = \int ds'_1 R_1(s'_1)E_{1O}(\mathbf{s}_{1O}) \exp(ik\mathbf{r}_{1H}(\mathbf{s}_{1O}) \cdot \mathbf{r}_{1H})$, where $R_1(s'_1)$ is the reflectivity coefficient of the magnifier, $\mathbf{r}_{1H}(\mathbf{s}_{1O})$ is implicitly defined by $\mathbf{n}_1 \times (\mathbf{r}_{1O} + \mathbf{H}/k - \mathbf{r}_{1H}) = 0$, and $\mathbf{H}$ is the Bragg vector. The corresponding angular spectrum is $E_{1H}(s'_{1H}) = J(s'_{1O}, s'_{1O})R_1(s'_1)E_{1O}(s_{1O})$, where $J(s'_{1O}, s'_{1O})$ is the Jacobian of $s'_{1O}$ with respect to $s'_{1H}$.

Assuming, for simplicity of discussion, the magnifier has negligible absorption, which is a good approximation for Si above about 15 keV,

$$R_1(s'_{1O})/R(0) = \begin{cases} \exp(i \arcsin(2x_{1O}/\omega_1)) & \text{if } |x'_{1O}| < \omega_1/2 \\ 2ix_{1O}/\omega_1 - i \text{sgn}(x_{1O})\sqrt{(2x_{1O}/\omega_1)^2 - 1} & \text{otherwise} \end{cases}$$

to first order in $s'_{1O}$, where $R(0) = i\sqrt{\psi_H/\psi_{1H}}/m$, $\psi_H$ and $\psi_{1H}$ are the Fourier coefficients of the magnifier’s dielectric constant at reciprocal lattice vectors $\mathbf{H}$ and $-\mathbf{H}$ respectively, $\omega_1$ is the magnifier’s rocking curve width, and
at 18 keV, and 50% at 27 keV. 60% just below the K absorption edge at 11.1 keV, 25% at 1700 Å, at wavelengths below about 10 Å. For the electron microscope, the transfer function

\[
\text{OTF}(s_o') = \frac{\exp(iks_d' \cdot s_{d'})}{s_{d'}},
\]

where \(s_o'\) is the spatial frequency in the object plane, \(s_{d'}\) is the spatial frequency in the detector plane, and \(k\) is the wave number. For optical microscopy, the transfer function

\[
\text{OTF}(s_o') = \frac{\exp(iks_d' \cdot s_{d'})}{s_{d'}},
\]

where \(s_o'\) is the spatial frequency in the object plane, \(s_{d'}\) is the spatial frequency in the detector plane, and \(k\) is the wave number. For electron microscopy, the transfer function

\[
\text{OTF}(s_o') = \frac{\exp(iks_d' \cdot s_{d'})}{s_{d'}},
\]

where \(s_o'\) is the spatial frequency in the object plane, \(s_{d'}\) is the spatial frequency in the detector plane, and \(k\) is the wave number. For x-ray microscopy, the transfer function

\[
\text{OTF}(s_o') = \frac{\exp(iks_d' \cdot s_{d'})}{s_{d'}},
\]

where \(s_o'\) is the spatial frequency in the object plane, \(s_{d'}\) is the spatial frequency in the detector plane, and \(k\) is the wave number.
degrade it. For illumination by a synchrotron radiation source with a crystal monochromator, the aberrations depend on the horizontal and vertical source sizes $X_s$ and $Y_s$, and the monochromator rocking curve width $\omega_m$. For the purpose of estimating typical aberrations, it is assumed that $X_s = 700 \, \mu m$, $Y_s = 40 \, \mu m$, the distance from the source to the object $Z_{so}$ is 60 m, the object is close to the first magnifier, $m \gg 1$ (e.g., $m = m_{opt}$), the magnifiers are just long enough to fully illuminate the detector, and the detector is square with length $L_d$ of 1 cm. The aberration due to the monochromator rocking curve width is roughly $L_d \omega_m/m \sin 2\theta_B$, which is negligible for a monochromator with Si (111) symmetric reflections. The aberration due to the vertical source size is roughly $L_d Y_s/Z_{so} \sin 2\theta_B$, which is also negligible. The aberration due to the horizontal source size is roughly $L_d X_s/Z_{so} \sin 2\theta_B$, which is $0.3 \, \mu m$ at $\lambda = 1 \, \AA$ for Si or Ge (111) reflections, and varies nearly as $\lambda^{-1}$ at shorter wavelengths. This aberration can be reduced by using a slit to effectively decrease $X_s$, and/or by using (220) reflections instead of (111).

In conclusion, the asymmetric Bragg diffraction microscope can form magnified in-line near-field holograms at submicrometer resolution, with high magnification and high efficiency, over a wide range of hard x-ray wavelengths. Its optical transfer function can be accurately computed from basic theory, because the magnifiers are nearly perfect crystals. As a result, absorption and phase contrast images can be computed from holograms acquired at several object distances. These characteristics make the microscope very promising for performing submicrometer resolution hard x-ray microtomography with synchrotron radiation.

[5] A crystal used in Bragg diffraction is said to be asymmetrically cut when its incident surface is inclined to the Bragg planes, and the diffraction is said to be asymmetric.
[10] Diffraction is called coplanar when the normal to the incident surface is coplanar with the incident beam wave vector and Bragg vector.
[11] The plane of diffraction is the plane containing the incident beam wave vector and Bragg vector.