Supplemental Materials for

Unveiling Nano-scale Crystal Deformation using Coherent X-ray Dynamical Diffraction

Longlong Wu[1,2*], David Yang[2], Wei Wang[2,3], Shinjae Yoo[4], Ross J. Harder[5], Wonsuk Cha[5], Aiguo Li[1] and Ian K. Robinson[2,6,‡]

[1]*Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201210, China*

[2]*Condensed Matter Physics and Materials Science Division, Brookhaven National Laboratory, Upton, New York 11973, USA*

[3]*Department of Physics, University of Science and Technology of China, 230026 Hefei, Anhui, China*

[4]*Computational Science Initiative, Brookhaven National Laboratory, Upton, NY 11973, USA*

[5]*Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois 60439, USA*

[6]*London Centre for Nanotechnology, University College London, London WC1H 0AH, UK*

Correspondence emails: [*]wull@sari.ac.cn, [†]irobinson@bnl.gov

Dynamical Theory:

For two-beam diffraction conditions, we get:

$$\mathbf{E}(\mathbf{r}) = E_0(\mathbf{r})e^{i\mathbf{k_0}\cdot\mathbf{r}} + E_h(\mathbf{r})e^{i\mathbf{k_h}\cdot\mathbf{r}}, \tag{S1}$$

where $\mathbf{k_0}$ and $\mathbf{k_h}$ represent the wavevectors of the transmitted and diffracted waves, respectively.

The complex wavefronts for the transmitted $E_0(\mathbf{r})$ and diffracted $E_h(\mathbf{r})$ waves are the solutions of the well-known Takagi-Taupin Equations (TTEs)[1-3]. For Bragg coherent X-ray diffraction imaging, they can be obtained by solving the symmetric version of two beam TTEs, given as

$$2i(\mathbf{k_0}\cdot\nabla)E_0(\mathbf{r}) = k^2\left[\chi_0 E_0(\mathbf{r}) + C\chi_{\bar{h}}E_h(\mathbf{r})e^{-i\Delta\mathbf{q}\cdot\mathbf{r}+i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})}\right],$$

$$2i(\mathbf{k_h}\cdot\nabla)E_h(\mathbf{r}) = k^2\left[\chi_0 E_h(\mathbf{r}) + C\chi_h E_0(\mathbf{r})e^{i\Delta\mathbf{q}\cdot\mathbf{r}-i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})}\right]. \tag{S2}$$

Here, χ_0, χ_h and $\chi_{\bar{h}}$ are Fourier coefficients of the susceptibility function of the crystal for the transmitted beam and diffracted beam, respectively. Here, $\mathbf{Q} = \mathbf{k_f} - \mathbf{k_0}$, $\mathbf{q} = \mathbf{Q} - \mathbf{h}$ and $\Delta\mathbf{q} = \mathbf{h}' - \mathbf{h}$. By defining an orthogonal grid with the three orthonormal unit vectors $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$, in a Bragg coherent X-ray diffraction imaging experiment, it can be generally assumed that $\mathbf{k_0}\cdot\hat{\mathbf{z}} > 0$ and $\mathbf{k}_h\cdot\hat{\mathbf{z}} > 0$. Thus, the two equations above can be reduced to:

$$2k_{0,z}\frac{\partial}{\partial z}E_0(\mathbf{r}) = -2(\mathbf{k_{0,\perp}}\cdot\nabla_\perp)E_0(\mathbf{r}) - ik^2\chi_0 E_0(\mathbf{r}) - ik^2 C\chi_{\bar{h}}E_h(\mathbf{r})e^{-i\Delta\mathbf{q}\cdot\mathbf{r}+i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})},$$

$$2k_{h,z}\frac{\partial}{\partial z}E_h(\mathbf{r}) = -2(\mathbf{k_{h,\perp}}\cdot\nabla_\perp)E_h(\mathbf{r}) - ik^2\chi_0 E_h(\mathbf{r}) - ik^2 C\chi_h E_0(\mathbf{r})e^{i\Delta\mathbf{q}\cdot\mathbf{r}-i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})}. \tag{S3}$$

Here, $\mathbf{k_0} = \mathbf{k_{0,z}} + \mathbf{k_{0,\perp}}$, $\mathbf{k_h} = \mathbf{k_{h,z}} + \mathbf{k_{h,\perp}}$, and $\nabla_\perp = [\frac{\partial}{\partial x}, \frac{\partial}{\partial y}]$. Therefore, they can be further simplified as

$$\frac{\partial}{\partial z}E_0(\mathbf{r}) = -\frac{ik^2}{2k_{0,z}}\chi_0 E_0(\mathbf{r}) - \frac{1}{k_{0,z}}(\mathbf{k_{0,\perp}}\cdot\nabla_\perp)E_0(\mathbf{r}) - \frac{ik^2}{2k_{0,z}}C\chi_{\bar{h}}E_h(\mathbf{r})e^{-i\Delta\mathbf{q}\cdot\mathbf{r}+i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})},$$

$$\frac{\partial}{\partial z}E_h(\mathbf{r}) = -\frac{ik^2}{2k_{h,z}}\chi_0 E_h(\mathbf{r}) - \frac{1}{k_{h,z}}(\mathbf{k_{h,\perp}}\cdot\nabla_\perp)E_h(\mathbf{r}) - \frac{ik^2}{2k_{h,z}}C\chi_h E_0(\mathbf{r})e^{+i\Delta\mathbf{q}\cdot\mathbf{r}-i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})}. \tag{S4}$$

Further, based on the differential property of Fourier transform, *i.e.,* $\mathcal{F}\left\{\frac{d}{dx}f(x)\right\} = i2\pi\xi\hat{f}(\xi)$, by applying the transverse Fourier transform into the two equations above, they can be rewritten as:

$$\frac{\partial}{\partial z}\tilde{E}_0(\boldsymbol{q}_\perp, z) = -\frac{i2\pi}{k_{0,z}}\mathbf{q}_\perp \cdot \mathbf{k}_{0,\perp} \times \tilde{E}_0(\boldsymbol{q}_\perp, z) - \frac{ik^2}{2k_{0,z}}\mathcal{F}_\perp\{\chi_{\bar{h}}E_h(\mathbf{r})e^{-i\Delta\mathbf{q}\cdot\mathbf{r}+i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})} + \chi_0 E_0(\mathbf{r})\},$$

$$\frac{\partial}{\partial z}\tilde{E}_h(\boldsymbol{q}_\perp, z) = -\frac{i2\pi}{k_{h,z}}\mathbf{q}_\perp \cdot \mathbf{k}_{h,\perp} \times \tilde{E}_h(\boldsymbol{q}_\perp, z) - \frac{ik^2}{2k_{h,z}}\mathcal{F}_\perp\{\chi_h E_0(\mathbf{r})e^{i\Delta\mathbf{q}\cdot\mathbf{r}-i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})} + \chi_0 E_h(\mathbf{r})\}. \quad \text{(S5)}$$

Here, $\mathcal{F}_\perp\{E(\mathbf{r}_\perp, z)\} = \int E(\mathbf{r}_\perp, z)e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}d\mathbf{r}$ and $\mathcal{F}_\perp^{-1}\{\tilde{E}(\mathbf{q}_\perp, z)\} = \int \tilde{E}(\mathbf{q}_\perp, z)e^{-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}d\mathbf{q}_\perp$. $\mathbf{q}_\perp = [q_x, q_y]$ and $\mathbf{q} = \mathbf{q}_\perp + \mathbf{q}_z$. Then, the equations (S5) can be rewritten to the form used by exponential Runge-Kutta methods:

$$\frac{\partial}{\partial z}\mathbf{E}(z) = A\mathbf{E}(z) + \mathbf{B}[z, \mathbf{E}(z)], \quad \text{(S6)}$$

where A is a diagonal matrix with $A = \begin{bmatrix} A_0 \\ A_h \end{bmatrix} = \begin{bmatrix} -\frac{i2\pi}{k_{0,z}}\mathbf{q}_\perp \cdot \mathbf{k}_{0,\perp} \\ -\frac{i2\pi}{k_{h,z}}\mathbf{q}_\perp \cdot \mathbf{k}_{0,\perp} \end{bmatrix}$. $\mathbf{E}(z) = \begin{bmatrix} \tilde{E}_0(\boldsymbol{q}_\perp, z) \\ \tilde{E}_h(\boldsymbol{q}_\perp, z) \end{bmatrix}$ and

$$\mathbf{B}(z, \mathbf{E}) = \begin{bmatrix} -\frac{ik^2}{2k_{0,z}}\mathcal{F}_\perp\{\mathcal{F}_\perp^{-1}(\tilde{E}_h)\chi_{\bar{h}}e^{-i\Delta\mathbf{q}\cdot\mathbf{r}+i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})} + \chi_0 E_0(\mathbf{r})\} \\ -\frac{ik^2}{2k_{h,z}}\mathcal{F}_\perp\{\mathcal{F}_\perp^{-1}(\tilde{E}_0)\chi_h e^{i\Delta\mathbf{q}\cdot\mathbf{r}-i\mathbf{h}\cdot\mathbf{u}(\mathbf{r})} + \chi_0 E_h(\mathbf{r})\} \end{bmatrix}$$. Eq. (S6) can be solved by

exponential Runge-Kutta methods of type given by Hochbruck and Ostermann[4]. Using the second exponential integrator, which is an explicit second-order method based on Heun's method, the solution for Eq. (S6) is given by the steps:

$$E_1^* = \mathbf{E}(z),$$
$$b_1 = \mathbf{B}(z_n, E_1^*)$$
$$E_2^* = \phi_0 E_1^* + h\phi_1 b_1,$$
$$b_2 = \mathbf{B}(z_n + h, E_2^*),$$

3

$$\mathbf{E}(z+h) = \phi_0 E_1^* + \frac{h}{2}[(2\phi_1 - \phi_2)b_1 + \phi_2 b_2], \tag{S7}$$

where, $\phi_0 = e^{hA}$ and $\phi_n = n(hA)^{-1}(\phi_{n-1} - 1)$.

With the abovementioned finite difference scheme, the TTEs can be solved on an arbitrary orthogonal grid and there are many different choices for the selection of the orthogonal grid. Since the main goal for BCDI here is to calculate the diffracted wavefront and demonstrate the dynamical effect, the origin of the coordinate system is chosen based on the scattering plane, aligned within the XOZ-plane, where $\mathbf{k}_h$ is along the z-axis. Thus, using this coordinate system, we get $\mathbf{k}_0 = [-k\sin(2\theta_B), 0, k\cos(2\theta_B)]$ and $\mathbf{k}_h = [0, 0, k]$. The relation between the laboratory coordinate X'Y'Z' and this coordinate XYZ is a rotation transformation, *i.e.*, $R(\delta, \gamma)\begin{bmatrix} x' \\ y' \\ z' \end{bmatrix} = \begin{bmatrix} x \\ y \\ z \end{bmatrix}$.

Kinematical approximation:

Here, we derive an analytical solution of the diffraction wavefront based on the kinematical approximation. In the kinematical approximation, the 3D diffraction signal is given as[5]:

$$A(\mathbf{q}) = \int s_h(\mathbf{r})e^{i\varphi_h(\mathbf{r})}e^{i\mathbf{q}\cdot\mathbf{r}} \, d\mathbf{r}, \tag{S8}$$

For Eq. S8, we assume the origin of the coordinate system is set based on the scattering plane, aligned within the XOZ-plane, where $\mathbf{k}_h$ is along the z-axis. In this coordinate system, the diffracted exit wavefront is perpendicular to the $\mathbf{k}_h$. To get the 2D diffraction wavefront at Δq_z, based on the Eq. S8, we get

$$A(\Delta q_z) = \int s_h(\mathbf{r})e^{i\varphi_h(r)}e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp + i\Delta q_z\cdot z} \, d\mathbf{r}$$

$$= \int\int s_h(\mathbf{r})e^{i\varphi_h(\mathbf{r})} e^{i\Delta q_z\cdot z} \, dz e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp} d\mathbf{r}_\perp, \tag{S9}$$

By further considering the shift of the exit wavefront by $\Delta\mathbf{q}_\perp$, Eq. S9 can be rewritten as

$$A(\Delta q_z, \Delta\mathbf{q}_\perp) = \int\int s_h(\mathbf{r})e^{i\varphi_h(\mathbf{r})+i\Delta q_z\cdot z} \, dz e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp + i\Delta\mathbf{q}_\perp\cdot\mathbf{r}_\perp} d\mathbf{r}_\perp$$

$$= \int\int s_h(\mathbf{r})e^{i\Delta\mathbf{q}\cdot\mathbf{r}+i\varphi_h(\mathbf{r})} \, dz e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp} d\mathbf{r}_\perp$$

$$= \int E(\Delta\mathbf{q}) e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp} d\mathbf{r}_\perp, \tag{S10}$$

Thus, the exit diffraction wavefront under kinematical case is given as $E(\Delta \mathbf{q}) = \int s_h(\mathbf{r}) e^{i\Delta \mathbf{q} \cdot \mathbf{r} + i\varphi(\mathbf{r})} \, dz$.
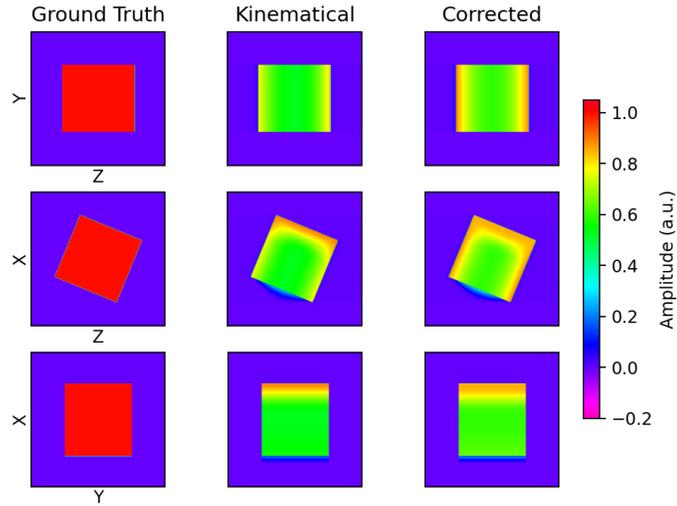
Supplemental Figure

Figure S1. Distortions appearing in the amplitude reconstruction of the simulated dynamical diffraction pattern of a strained gold cube using the traditional (kinematical) approach and the result of correcting them for absorption. The first column shows the central slices of the amplitude used for simulation. The middle column shows the corresponding reconstructed results from the traditional iterative method. The last column shows the central slices of the corrected amplitude.
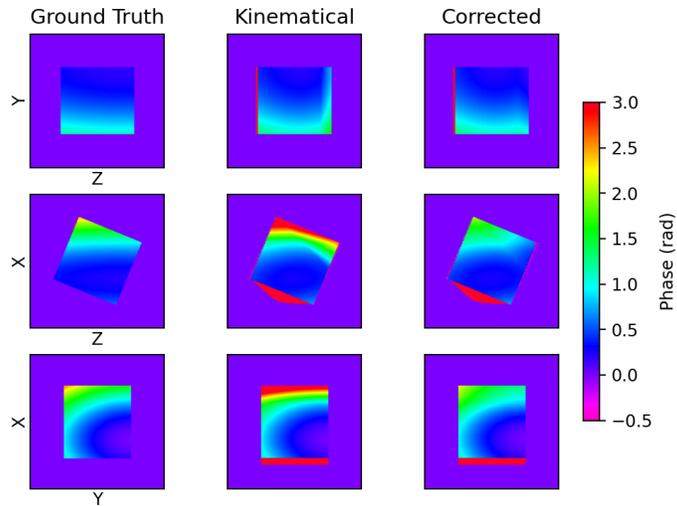

Figure S2. Distortions appearing in the phase reconstruction of the simulated dynamical diffraction pattern of a strained gold cube using the traditional (kinematical) approach and the result of correcting them for refraction. The first column shows the central slices of the phase used for simulation, showing the small strain introduced. The middle

column shows the corresponding reconstructed phase from the traditional iterative method. The last column shows the central slices of the corrected phase.

Supplemental References

1   Takagi, S. A Dynamical Theory of Diffraction for a Distorted Crystal. *J. Phys. Soc. Jpn.* **26**, 1239–&, (1969).
2   Taupin, D. Prevision De Quelques Images De Dislocations Par Transmission Des Rayons X (Cas De Laue Symetrique). *Acta Crystallogr.* **23**, 25–&, (1967).
3   Takagi, S. Dynamical Theory of Diffraction Applicable to Crystals with Any Kind of Small Distortion. *Acta Crystallogr.* **15**, 1311–&, (1962).
4   Friedli, A. Numerical Treatment of Differential Equations (Proc. Conf. Math. Forschungsinst. Oberwolfach, 1976). *Lecture Notes in Mathematics* **631**, 35–50, (1978).
5   Robinson, I. K., Vartanyants, I. A., Williams, G. J., Pfeifer, M. A. & Pitney, J. A. Reconstruction of the shapes of gold nanocrystals using coherent x-ray diffraction. *Phys. Rev. Lett.* **87**, 195505, (2001).