Supplementary Information

Nanoscale Ultrafast Lattice Modulation with Hard X-ray Free Electron Laser

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Supplementary Note 1 Experimental Setup

Note 1.1 Ray tracing analysis

The synchronization of the two pump pulses on the sample is achieved through the symmetry of the trajectory within the x-z plane as shown in Extended Data Figure 1(b). Due to installation, alignment, and motor operation errors, M_1 and M_2 are not ideally symmetric. We use ray-tracing analysis to estimate the tolerance of installation and alignment errors of M_1 and M_2.

Extended Data Figure 2(a) shows the X-ray trajectory between G and the sample. In the experimental setup, $d_0 \approx 6$ m. The alignment uncertainty of the X-ray in the transverse plane is assumed to be $\Delta\eta \leq 2$ μm. The coherence time of the X-ray pulse after the split-delay optics is 10 fs. To guarantee a high contrast of the interference between the two X-ray pulses, the path length difference should be smaller than 10% of the coherence length, i.e., $|\Delta(l_1 + l_2)| \leq 0.3$ μm.

For a given grating diffraction angle $\alpha_0/2$, grating-sample distance d_0, and the beam position on the sample plane $\eta = 0$, the total path length $l_1 + l_2$ is solely determined by the mirror angle α. It can be shown that:

$$l_1 + l_2 = \frac{d_0 \left(\sin \frac{\alpha_0}{2} + \sin \bar{\alpha}\right) + v \left(\cos \bar{\alpha} - \cos \frac{\alpha_0}{2}\right)}{\sin(2\alpha)}. \tag{SI-1}$$

where $\bar{\alpha} \equiv 2\alpha - \frac{\alpha_0}{2}$.

By uniformly sampling $\eta \in [-2, 2]$ μm and $\alpha \in [0.15° - 0.1°, 0.15° + 0.1°]$, and assuming that $d_0 = 6$ m, the range of the deviation of $l_1 + l_2$ from its ideal value at $\eta = 0$ and $\alpha = 0.15°$ is shown as a function of $\Delta\alpha$ in Extended Data Figure 2(b). In Extended Data Figure 2(b), green vertical lines indicate $\Delta\alpha = \pm 0.01°$, while the red horizontal lines indicate $\Delta(l_1 + l_2) = \pm 0.3$ μm. Therefore, to achieve sub-femtosecond synchronization, we only need to guarantee that $|\Delta\eta| \leq 2$ μm and that $|\Delta\alpha| \leq 0.01°$.

Note 1.2 Mirror angle calibration

In this experiment, we control mirror angles with motorized stages. A stage with model SA10A-RT01 (Kohzu Precision Co.,Ltd.) is used to rotate M_0, while two stages with model SA05A-RT02 (Kohzu Precision Co.,Ltd.) are used for M_1 and M_2. The angle accuracy of SA05A-RT02 (Kohzu Precision Co.,Ltd.) is 0.000765° according to the spec, significantly smaller than the 0.01° requirement. Therefore, the angle uncertainty of M_1 and M_2 is dominated by the installation and alignment accuracy.

Denote the feedback value of the rotation stage with α_f. The α_f has an offset with respect to the grazing angle α with respect to the incident X-ray pulse: $\alpha_f \equiv \alpha + \alpha_{\text{off}}$, where α_{off} indicates the offset value. Due to the high motor motion accuracy, we assume the uncertainty of α to equal that of α_{off}. The uncertainty of α_{off} is determined by analyzing the X-ray profile on the sample plane beam profile monitor with partial reflections as shown in Extended Data Figure 3(a).

In Extended Data Figure 3(a), the distance between the rotation center and beam center is l_0, the beam size is L, and the distance between the rotation center and the sample plane is D. Assume that, without the mirror, the lower edge of the beam is at location O. The mirror length is l_M and we assume that the rotation center of the mirror is at the center of the reflection surface. For a given grazing angle α, the lower edge of the X-ray that passes by the mirror is at a distance of s_1 with respect to O, while the upper edge of the reflected pulse has a distance s_2 with respect to O. We can show that:

$$s_1 = \frac{l_M}{2}\sin\alpha - \left(l_0 - \frac{L}{2}\right), \tag{SI-2}$$

$$s_2 = \left(D - \left(l_0 - \frac{L}{2}\right)\frac{1}{\tan\alpha}\right)\tan(2\alpha). \tag{SI-3}$$

Therefore,

$$s_2 - s_1 = D\tan(2\alpha) - \left(l_0 - \frac{L}{2}\right)\left(\frac{\tan(2\alpha)}{\tan\alpha} - 1\right) - \frac{l_M}{2}\sin\alpha. \tag{SI-4}$$

We align the mirror rotation center to the lower edge of the incident X-ray pulse and therefore $l_0 \approx L/2$. Because $D \approx 0.1\ m \gg l_0 \approx 75\ \mu m$, $D \gg L \approx 150\mu m$ and $l_M/2 \approx 0.02m$,

$$S_2 - S_1 \approx\ D\tan(2\alpha_f - 2\alpha_{\mathrm{off}}). \tag{SI-5}$$

For a series of α_f as documented in Extended Data Table 1 for M_1 and M_2, we measure the edge separation $S_2 - S_1$.

We then compute the mean error, $\Xi(D, \alpha_{\mathrm{off}})$, between the measured edge separation $S_2 - S_1$ and that from the SI-5 for a given D and α_{off}. Here, the mean error, $\Xi(D, \alpha_{\mathrm{off}})$, is defined as:

$$\Xi(D, \alpha_{off}) \equiv \sqrt{\frac{1}{n}\sum_{l=1}^{n}\left(S'(\alpha_{f,l};\ Exp) - S'(\alpha_{f,l};\ \alpha_{off}, D)\right)^2}, \tag{SI-6}$$

$$S'(\alpha_f;\ \alpha_{\mathrm{off}}, D) \equiv\ D\tan(2\alpha_f - 2\alpha_{\mathrm{off}}), \tag{SI-7}$$

$$S'(\alpha_f;\ Exp):\ \text{Measured } S_2 - S_1 \text{ at } \alpha_f. \tag{SI-8}$$

Here, n is the total measurement number and $\alpha_{f,l}$ indicates the l^{th} motor feedback value.

The value of $\Xi(D, \alpha_{\mathrm{off}})$ for a range of D and α_{off} is shown in Extended Data Figure 3(c) and (d) respectively for the measurement of M_1 and M_2. The position of the global minimum of $\Xi(D, \alpha_{\mathrm{off}})$ gives the nominal D and α_{off} for us to improve the symmetry of the X-ray trajectory for the two pump pulses. The purple ellipse in Extended Data Figure 3(c) and (d) encloses the region smaller than twice of the global minimum of $\Xi(D, \alpha_{\mathrm{off}})$. The extension of the projection of the purple ellipse along the axes determines the uncertainty of D and α_{off}, respectively. For both M_1 and M_2, the uncertainty in α_{off} is 0.003°. This results in the 0.003° uncertainty of α utilized in this work.

Note 1.3 Detector geometry calibration

The scattered wave-vector Q for each pixel on our detector is calibrated with the powder rings from a LaB_6 powder sample. The LaB_6 sample is installed on the sample mount for STO. Through the optimization of the overlap of the documented ring position with measured powder rings as shown in Extended Data Figure 8, detector distance is determined to be 12 cm downstream the X-ray-sample interaction point along the z axis, and the lower right corner of the detector is 89.2 mm and -11.0 mm away from the X-ray sample interaction point along the x and y axis. The Extended Data Figure 8 (b), (c) and (d) show the overlap of the measured powder ring with documented ring positions with calibrated detector position and distance.

In the analysis, only a small region around the Bragg peak is of interest. The Q of each pixel within this region is shwon in Extended Data Figure 9 (a) where the number on the axes indicates the corresponding pixel index in Extended Data Figure 8 (a). The equivalent reduced wave-vector in the first Brillouin zone, q, for each pixel is shown in Extended Data Figure 9 (b), (c) and (d) for different XTG measurements. The text in the lower left corner of Extended Data Figure 9 of (b), (c) and (d) indicates Λ_{TG} and the transient grating wave-vector.

Due to the high angle accuracy of M_1 and M_2, we use the XTG wave-vector as the reference wave-vector to derive q for other pixels. In the derivation, we first derive $\mathbf{Q}$ for each pixel, and then derive $\mathbf{H}$ according to $\pm\mathbf{k}_{\mathrm{TG}} = \mathbf{q} \equiv \mathbf{Q} - \mathbf{H}$ with the $\mathbf{Q}$ of the pixel on the XTG peak. With $\mathbf{H}$, we obtain the $\mathbf{q}$ for other pixels shown in Extended Data Figure 9.

Supplementary Note 2 Transient Grating Theory

Note 2.1 Scattering intensity from lattice modification

We use kinematic X-ray scattering theory to analyze the measured signal. The dominant signal observed in the measurement comes from the longitudinal acoustic phonon. Therefore, we assume that the unit cell of the $SrTiO_3$ (STO) does not change during evolution.

According to kinematic X-ray scattering theory, the scattering intensity at wavevector $\mathbf{Q}$ is:

$$I(\mathbf{Q},\tau) = \Phi r_e^2 P^2 \Omega_p \left\langle \left| \sum_l F(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{R}_l(\tau)} \right|^2 \right\rangle, \tag{SI-9}$$

where, r_e is the classical electron radius, P is the polarization factor, Φ is the incident X-ray photon flux per pulse, and Ω_p is the solid angle of the detector pixel. In addition, $F(\mathbf{Q})$ is the STO unit cell form factor, $\mathbf{R}_l(\tau)$ is the position of the l^{th} unit cell in the illumination volume and the summation is taken over the illumination volume. The $\langle\cdot\rangle$ indicates the averaging over different X-ray pulses. In the following, we define $I_{th} \equiv \Phi r_e^2 P^2 \Omega_p$.

Assume that the coherently scattering volume is V, and the total illumination volume is V'. Assume that $\mathbf{R}_l(\tau) \equiv \mathbf{R}_l + \mathbf{R}'_l(\tau) + \mathbf{U}_l(\tau)$, where $\mathbf{R}_l$ is the ideal lattice position, $\mathbf{R}'_l(\tau)$ represents thermal fluctuations and disorders that lead to the equilibrium diffuse scattering signal without pump X-ray pulses, and $\mathbf{U}_l(\tau)$ is the XTG induced lattice deformation field. Then the scattering intensity is:

$$\begin{aligned}
I(\mathbf{Q},\tau) \approx & I_{th}\frac{V'}{V}\left\langle \left| \sum_l F(\mathbf{Q}) e^{-i\mathbf{Q}\cdot\left(\mathbf{R}_l+\mathbf{R}'_l(\tau)\right)} + F(\mathbf{Q}) e^{-i\mathbf{Q}\cdot\mathbf{R}_l} i\mathbf{Q}\cdot\mathbf{U}_l(\tau) \right|^2 \right\rangle_V \\
=& I_0(\mathbf{Q}) + I_{th}\left|F(\mathbf{H})\right|^2 \frac{V'}{V_c^2 V}\left\langle \left| \sum_l e^{-i(\mathbf{Q}-\mathbf{H})\cdot\mathbf{R}_l}\mathbf{Q}\cdot\mathbf{U}_l(\tau) V_c \right|^2 \right\rangle_V \\
\approx & I_0(\mathbf{Q}) + I_{th}\left|F(\mathbf{H})\right|^2 \frac{V'}{V_c^2 V}\left\langle \left| \int_V e^{-i(\mathbf{Q}-\mathbf{H})\cdot\mathbf{R}}\mathbf{Q}\cdot\mathbf{U}(\mathbf{R},\tau)\, d^3\mathbf{R} \right|^2 \right\rangle_V \\
=& I_0(\mathbf{Q}) + I_{th}\left|F(\mathbf{H})\right|^2 \frac{V'}{V_c^2 V}\left\langle \left| \mathbf{Q}\cdot\widetilde{\mathbf{U}}(\mathbf{Q}-\mathbf{H},\tau) \right|^2 \right\rangle_V .
\end{aligned} \tag{SI-10}$$

Here, V_c is the unit cell volume, $I_0(\mathbf{Q})$ is the equilibrium X-ray scattering signal without the pump pulse and that,

$$\mathbf{Q}\cdot\widetilde{\mathbf{U}}(\mathbf{Q}-\mathbf{H},\tau) \equiv \int_V e^{-i(\mathbf{Q}-\mathbf{H})\cdot\mathbf{R}}\mathbf{Q}\cdot\mathbf{U}(\mathbf{R},\tau)\, d^3\mathbf{R}. \tag{SI-11}$$

The $\langle\cdot\rangle_V$ indicates the averaging over different V within V' and averaging over different X-ray pulses, and we assume that $\mathbf{H}$ is the only reciprocal lattice vector of the sample that is close to $\mathbf{Q}$, which is suitable for our measurement.

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Note 2.2 Strain field of single photon ionization

Due to the small cross section, each absorbed X-ray photon creates its own displacement field $\mathbf{u}(\mathbf{r},0)$ around the absorption location. We assume that the absorption of an X-ray photon leads to a localized spherically symmetric distortion in the equilibrium position of atoms at time $\tau = 0$, which launches longitudinal spherical waves for $\tau > 0$ and that the distortion field is curl-free. Therefore, it is the gradient of the auxiliary field ϕ:

$$\mathbf{u}\left(\mathbf{r},0\right) \equiv \nabla\phi\left(\mathbf{r},0\right). \tag{SI-12}$$

Following the same derivation as that shown in [1], the dynamics of the displacement field can be described with the following equation and initial conditions:

$$-\left(q^2 + \frac{2}{v^2\tau_1}\frac{\partial}{\partial\tau} + \frac{1}{v^2}\frac{\partial^2}{\partial\tau^2}\right)\widetilde{\phi}\left(\mathbf{q},\tau\right) = \widetilde{s}(\mathbf{q})e^{-\frac{\tau}{\tau_0}}\ H\left(\tau\right), \tag{SI-13}$$

$$\widetilde{\phi}\left(\mathbf{q},\tau\right)|_{\tau=0} = 0, \tag{SI-14}$$

$$\frac{\partial}{\partial\tau}\widetilde{\phi}\left(\mathbf{q},0\right) = 0, \tag{SI-15}$$

$$\widetilde{s}\left(\mathbf{q}\right) = u_0 \exp\left(-\frac{1}{2}\sigma^2 q^2\right), \tag{SI-16}$$

where $\widetilde{s}\left(\mathbf{q}\right)$ is the source of the distortion field in reciprocal space, $H\left(\tau\right)$ is the Heaviside step function, the $e^{-\frac{\tau}{\tau_0}}$ accounts for relaxation of the distortion of the equilibrium position, and that the $\frac{2}{v^2\tau_1}\frac{\partial}{\partial\tau}$ accounts for phonon relaxation due to various phonon scattering processes. The constant value v is the sound velocity of the longitudinal acoustic phonon mode in STO. The τ_0 and τ_1 are fitted from the measurement data and are less than 10 ps. The functional form of $\widetilde{s}\left(\mathbf{q}\right)$ assumes that around each X-ray photon absorption site, the source of the distortion in the equilibrium position follows a Gaussian distribution with an amplitude u_0 and size σ.

The exact solution of this equation is:

$$\widetilde{\phi}\left(\mathbf{q},\tau\right) = B\left(e^{-\frac{\tau}{\tau_0}} - e^{-\frac{\tau}{\tau_1}}\cos\left(f\tau\right) - \frac{\tau_1-\tau_0}{f\tau_0\tau_1}e^{-\frac{\tau}{\tau_1}}\sin\left(f\tau\right)\right). \tag{SI-17}$$

$$B = -\frac{u_0}{q^2 - 2\left(v^2\tau_1\tau_0\right)^{-1} + \left(v^2\tau_0^2\right)^{-1}}\exp\left(-\frac{1}{2}\sigma^2 q^2\right), \tag{SI-18}$$

$$f = qv\sqrt{1-\left(q^2v^2\tau_1^2\right)^{-1}}. \tag{SI-19}$$

$$\tag{SI-20}$$

We assume that:

$$q^2v^2\left(\min\left(\tau_1,\tau_0\right)\right)^2 \gg 1, \tag{SI-21}$$

$$qv\tau_1\tau_0 \gg \left|\tau_1-\tau_0\right|. \tag{SI-22}$$

The result is simplified to:

$$\widetilde{\phi}(\mathbf{q},\tau) \approx -\frac{u_0}{q^2}\exp\left(-\frac{1}{2}\sigma^2 q^2\right)\left(e^{-\frac{\tau}{\tau_0}} - e^{-\frac{\tau}{\tau_1}}\cos(qv\tau)\right). \quad \text{(SI-23)}$$

Therefore, the deformation associated with the longitudinal acoustic phonon with a wavevector $\mathbf{q}$ is:

$$\widetilde{\mathbf{u}}(\mathbf{q},\tau) = \mathbf{q}\widetilde{\phi}(\mathbf{q},\tau) = -\hat{\mathbf{q}}\frac{u_0}{q}\exp\left(-\frac{1}{2}\sigma^2 q^2\right)\left(e^{-\frac{\tau}{\tau_0}} - e^{-\frac{\tau}{\tau_1}}\cos(qv\tau)\right), \quad \text{(SI-24)}$$

where $\hat{\mathbf{q}} \equiv \frac{\mathbf{q}}{|\mathbf{q}|}$. This finishes the derivation of the strain field model for each single X-ray photon absorption used in this paper.

Here, the relation between $\widetilde{\mathbf{u}}(\mathbf{q},\tau)$ and the real space displacement field, $\mathbf{u}(\mathbf{r},\tau)$, is:

$$\widetilde{\mathbf{u}}(\mathbf{q},\tau) \equiv \int \exp(-i\mathbf{q}\cdot\mathbf{r})\,\mathbf{u}(\mathbf{r},\tau)d\mathbf{r}. \quad \text{(SI-25)}$$

Note 2.3 Coherent enhancement

Assume that on average there N single X-ray photon ionization sites in each coherent scattering volume V. The total displacement field is therefore:

$$\mathbf{U}(\mathbf{R},\tau) = \sum_{l}^{N}\mathbf{u}(\mathbf{R}-\mathbf{r}_l,\tau). \quad \text{(SI-26)}$$

Here, $\mathbf{r}_l$ is the location of the l^{th} photon ionization site. Therefore:

$$\widetilde{\mathbf{U}}(\mathbf{q},\tau) = \widetilde{\mathbf{u}}(\mathbf{q},\tau)\sum_{l}^{N}\mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}_l}. \quad \text{(SI-27)}$$

Therefore, the total scattering intensity is:

$$I(\mathbf{Q},\tau) = I_0(\mathbf{Q}) + I_{th}|F(\mathbf{H})|^2\frac{V'}{V_c^2 V}|\mathbf{Q}\cdot\widetilde{\mathbf{u}}(\mathbf{q},\tau)|^2\left\langle\left|\sum_{l}^{N}\mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}_l}\right|^2\right\rangle_V. \quad \text{(SI-28)}$$

For the l^{th} X-ray photon absorption event, the probability density to find it at location $\mathbf{r}_l$ is $P(\mathbf{r}_l) = V^{-1}(1+\beta\cos(\mathbf{r}_l\cdot\mathbf{k}_{TG}))$, where β is the contrast of the X-ray standing wave. In our sparse photon absorption event limit, we can consider the photon absorption events to be independent from each other. Therefore:

$$\left\langle\left|\sum_{l}^{N}\mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}_l}\right|^2\right\rangle_V = N + \sum_{l\neq m}^{N}\left\langle\mathbf{e}^{i\mathbf{q}\cdot(\mathbf{r}_l-\mathbf{r}_m)}\right\rangle_V$$

$$=N + N(N-1)\int_{V^2} \mathbf{e}^{i\mathbf{q}\cdot(\mathbf{r}_1-\mathbf{r}_2)} P(\mathbf{r}_1) P(\mathbf{r}_2) d^3\mathbf{r}_1 d^3\mathbf{r}_2$$
$$\approx N + N(N-1)\left(s(\mathbf{q}) + \frac{1}{4}\beta^2 s(\mathbf{q}-\mathbf{k}_{TG}) + \frac{1}{4}\beta^2 s(\mathbf{q}+\mathbf{k}_{TG})\right) \tag{SI-29}$$

Here,

$$s(\mathbf{q}) \equiv \left|\frac{1}{V}\int_V e^{i\mathbf{q}\cdot\mathbf{r}} d^3\mathbf{r}\right|^2. \tag{SI-30}$$

When V is sufficiently large, $s(\mathbf{q}) \approx \delta_{\mathbf{q},\mathbf{0}}$ where $\delta_{\mathbf{q},\mathbf{0}}$ is the Kronecker delta. Since $\mathbf{q} = \mathbf{Q} - \mathbf{H}$, when $\mathbf{q} = \mathbf{0}$, it means we are measuring the Bragg diffraction which is not the focus of this study. Therefore, we only consider $\mathbf{q} \neq \mathbf{0}$. In this case,

$$\left\langle \left|\sum_l^N \mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}_l}\right|^2 \right\rangle_V = \begin{cases} N & if\ \mathbf{q} \neq \mathbf{0}, \pm\mathbf{k}_{TG} \\ 1 + \beta^2 \frac{N(N-1)}{4} & if\ \mathbf{q} = \pm\mathbf{k}_{TG} \end{cases}. \tag{SI-31}$$

In our experiment, the calibration procedure guarantees a high contrast of the X-ray standing wave. Therefore, we assume $\beta = 1$. This yields the cited coherent enhancement factor in the main text. Combining with the solid angle comparison between the XTG peak and the pixel solid angle, we obtain the theoretical estimation of the overall enhancement with XTG excitation.

Note 2.4 Angular divergance

According to the kinematic diffraction theory, for a crystal of a thickness l and an X-ray with wavelength λ_0, the angular spread is [2]:

$$\Delta\phi' = 0.443\frac{\lambda_0}{l}\frac{1}{\cos\theta_B}. \tag{SI-32}$$

Here, θ_B is the Bragg angle. This $\Delta\phi'$ is in the diffraction plane spanned by $\mathbf{k}_3$, and the reciprocal lattice $\mathbf{H}$. Since we are rotating around the sample surface normal direction $\mathbf{n}$, we need to account for the rotation axis difference when comparing with the measured angular spread.

Define:

$$\sigma \equiv \frac{\mathbf{k}_3 \times \hat{\mathbf{H}}}{\left|\mathbf{k}_3 \times \hat{\mathbf{H}}\right|}, \tag{SI-33}$$
$$\hat{\mathbf{H}} \equiv \frac{\mathbf{H}}{|\mathbf{H}|}. \tag{SI-34}$$

For a rotation of $\mathbf{H}$ around σ of $\Delta\phi'$, the change of the reciprocal lattice is:

$$\mathbf{h}\prime \equiv (\sigma \times \mathbf{H})\,\Delta\phi'. \tag{SI-35}$$

For a rotation of $\mathbf{H}$ around $\mathbf{n}$ of $\Delta\phi$, the change of the reciprocal lattice is:

$$\mathbf{h} \equiv (\mathbf{n} \times \mathbf{H})\,\Delta\phi. \tag{SI-36}$$

The $\Delta\phi$ is what we measure in the experiment. We require the projection of $\mathbf{h}$ in the diffraction plane to be of the same length as $\mathbf{h}'$. Therefore:

$$|\mathbf{h} - (\sigma \cdot \mathbf{h})\,\sigma| = |\mathbf{h}'|\,, \tag{SI-37}$$

$$\Rightarrow \left(((\mathbf{n} \times \mathbf{H}))^2 - (\sigma \cdot ((\mathbf{n} \times \mathbf{H})))^2\right)(\Delta\phi)^2 = (\sigma \times \mathbf{H})^2\,(\Delta\phi')^2 = (\Delta\phi')^2\,. \tag{SI-38}$$

With vectorial algebra, one can prove that:

$$((\mathbf{n} \times \mathbf{H}))^2 - (\sigma \cdot ((\mathbf{n} \times \mathbf{H})))^2 = |\mathbf{n} \cdot \sigma|^{\mathbf{2}}\,. \tag{SI-39}$$

Therefore, define η to be the angle between $\mathbf{n}$ and σ:

$$\cos\eta \equiv \mathbf{n} \cdot \sigma \tag{SI-40}$$

Then:

$$\Delta\phi = \frac{\Delta\phi'}{\cos\eta} = 0.443\frac{\lambda_0}{l}\frac{1}{\cos\theta_B}\frac{1}{\cos\eta} \tag{SI-41}$$

In our measurement, l corresponds to the spatial coherent length of the periodic excitation from the X-ray interference pattern.

Supplementary Note 3 Data Analysis

Note 3.1 Bragg diffraction simulation

Vectors in Extended Data Figure 1(e) take the following value at $\phi = 0°$:

$$\mathbf{n} = (0.980, 0., -0.199), \tag{SI-42}$$

$$\mathbf{H} = (3.153, 3.218, -0.642)\ \text{Å}^{-1}, \tag{SI-43}$$

$$\mathbf{k}_1 = (0.026, 0.000, 4.966)\ \text{Å}^{-1}, \tag{SI-44}$$

$$\mathbf{k}_2 = (0.026, 0.000, 4.966)\ \text{Å}^{-1}, \tag{SI-45}$$

$$\mathbf{k}_3 = (-0.117, -1.973, 4.556)\ \text{Å}^{-1}, \tag{SI-46}$$

$$\mathbf{k}_{\text{TG}} = (0.053, 0., 0.)\ \text{Å}^{-1}. \tag{SI-47}$$

During the sample rotation around its normal direction $\mathbf{n}$, we calculate the reflectivity according to the two-beam dynamical diffraction theory, assuming a thick perfect crystal. The source code of the simulation is provided in the **Supplementary Data**.

Note 3.2 XTG-enhanced scattering intensity modulation

For the measured intensity modulation $r(\mathbf{q}, \tau)$ with $\Lambda_{\text{TG}} = 11.8$ nm, we find the first maxima of $r(\mathbf{q}, \tau)$ in τ for each q, which are marked with black dashed lines in Figure 2(c) and (d). Along the line of first maxima with each q, the value of $r(\mathbf{q}, \tau)$ is shown in Extended Data Figure 6(a) with solid lines. We perform a linear fit of the line cut, and the result is shown with dashed lines in Extended Data Figure 6(a).

The ratio between the measured intensity modulation maxima, $r_{\text{exp}} - 1$, versus the linear fit, $r_{\text{fit}} - 1$, is shown in Extended Data Figure 6(b). From Extended Data Figure 6(b), we determine that XTG induces an enhancement of the scattering intensity modulation at the XTG peak by a factor of 17 for $-\mathbf{k}_{\text{TG}}$ and 11 for $\mathbf{k}_{\text{TG}}$ in our measurement.

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