

Supplementary Information for

Soft-x-ray free-electron laser pulses stabilize a competing order in helimagnet

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I XFEL pulse monitoring and extraction of the resonant scattering signal

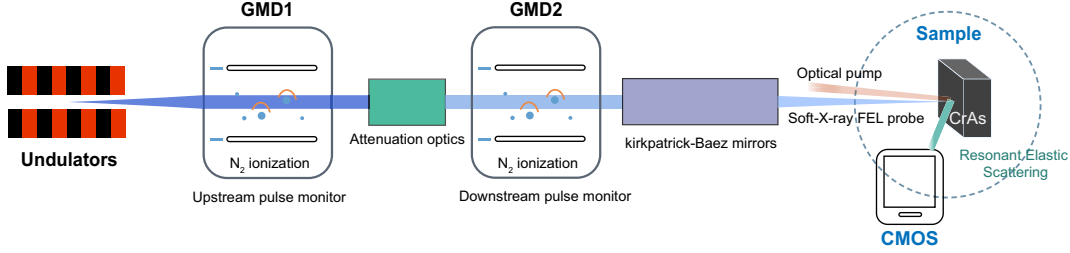


Figure S1: **XFEL pulse monitoring and resonant scattering detection.** The soft-x-ray FEL probe pulses were monitored by GMD1 before passing through attenuation optics, which reduced the pulse intensity to the level used in the experiment. The attenuated pulses were subsequently monitored by GMD2 and focused onto the CrAs sample using Kirkpatrick–Baez mirrors. As the downstream monitor, the GMD2 signal was used as the shot-by-shot reference for the incident XFEL probe intensity. At the sample position, the soft-x-ray probe spatially overlapped with the optical pump, and the resulting resonant elastic scattering signal was recorded using a CMOS area detector.

I.A XFEL beam transport and gas-monitor detector normalization

Figure S1 illustrates the beam transport, pulse monitoring, attenuation, focusing, and scattering-detection scheme used in the time-resolved resonant soft-x-ray scattering (tr-RSXS) experiment. The soft-x-ray FEL probe pulses were tuned near the Cr L_3 absorption edge and monitored by GMD1 before passing through attenuation optics, which reduced the pulse energy to the level used in the measurements. The attenuated pulses were subsequently monitored by GMD2 and then transported to the CrAs sample through a downstream Kirkpatrick–Baez optical system. Both GMDs measure the pulse intensity through the current generated by photoionization of dilute N_2 gas. Because GMD2 is located downstream of the attenuation optics and closer to the experimental endstation, its signal was used as the shot-by-shot reference for the incident XFEL probe intensity.

The incident XFEL photon number was estimated from the GMD signal. For a working gas with pressure p and temperature T , the molecular number density is $n = p/(k_B T)$. The collected GMD current can be expressed as

$$I_{\text{GMD}} = \frac{p}{k_B T} \dot{N}_\gamma \sigma_{\text{ph}}(\hbar\omega) z q_{\text{ph}}(\hbar\omega) e, \quad (\text{S1})$$

where $\dot{N}_\gamma$ is the incident photon rate, σ_{ph} is the photoionization cross section of the working gas, z is the effective interaction length, q_{ph} is the average charge produced per photoionization event, and e is the elementary charge. The photon number contained in one acquisition window is therefore

$$N_\gamma = \frac{I_{\text{GMD}} \Delta t k_B T}{\sigma_{\text{ph}} z p q_{\text{ph}} e}, \quad (\text{S2})$$

where Δt is the integration time of the GMD signal.

For the measurements reported here, the GMD was operated with N_2 gas at $T = 300$ K and $p = 1.0 \times 10^{-3}$ mbar. Using an effective interaction length $z = 200$ mm, a photoionization cross section $\sigma_{\text{ph}} = 0.647$ Mb near the Cr L_3 edge [1, 2], $q_{\text{ph}} = 1.256$, and an integration time $\Delta t = 4$ ms, a representative GMD2 signal $I_{\text{GMD2}} = 1.644$ nA gives

$$N_{\gamma, \text{GMD2}} \simeq 1.05 \times 10^{11} \text{ photons pulse}^{-1}. \quad (\text{S3})$$

Between GMD2 and the sample, the XFEL beam underwent six reflections in the downstream Kirkpatrick–Baez optical system. Taking the combined transmission of these reflections to be $\eta_{\text{KB}} = 0.262$, the photon number incident on the sample was estimated as

$$N_{\gamma, \text{sample}} = \eta_{\text{KB}} N_{\gamma, \text{GMD2}} \simeq 2.75 \times 10^{10} \text{ photons pulse}^{-1}. \quad (\text{S4})$$

Table S1: **XFEL irradiation parameters used to create the induced state.**

Parameter	Value
Repetition rate	10 Hz
Photons per pulse	2.75×10^{10}
Pulse duration	236 fs FWHM
X-ray footprint on sample	$57 \mu\text{m} \times 82 \mu\text{m}$ FWHM (44° incidence)
Accumulated exposure	$\sim 1.2 \times 10^4$ pulses
Peak photon fluence	$\sim 5.2 \times 10^{14}$ photons cm^{-2} pulse $^{-1}$
Peak photon flux density	2.1×10^{27} photons $\text{s}^{-1} \text{cm}^{-2}$
Sample temperature	29 K
Incident polarization	π polarization

I.B XFEL photon fluence and peak photon flux density

The incident XFEL beam had an approximately Gaussian spatial profile with a circular FWHM diameter of about $57 \mu\text{m}$. In the scattering geometry used to probe the $\mathbf{q}_2 \simeq (0, 0, 0.396)$ reflection, the x-ray incidence angle relative to the sample surface was approximately 44° . The beam footprint on the sample surface was therefore an ellipse with FWHM dimensions of approximately $57 \mu\text{m} \times 82 \mu\text{m}$. The FEL pulse duration was $\Delta t_X = 236$ fs (FWHM). The XFEL-induced $\mathbf{q}_2$ reflection became detectable before the sample had accumulated an exposure of approximately 1.2×10^4 FEL pulses.

For a pulse containing N_γ photons and an elliptical Gaussian beam footprint with FWHM dimensions d_x and d_y , the peak photon fluence is

$$F_{\gamma,\text{peak}} = \frac{4 \ln 2}{\pi d_x d_y} N_\gamma. \quad (\text{S5})$$

Using $N_\gamma = 2.75 \times 10^{10}$, $d_x = 57 \mu\text{m}$, and $d_y = 82 \mu\text{m}$, the peak photon fluence is

$$F_{\gamma,\text{peak}} \simeq 5.2 \times 10^{14} \text{ photons cm}^{-2} \text{ pulse}^{-1}. \quad (\text{S6})$$

Assuming a Gaussian temporal profile with FWHM duration Δt_X , the corresponding peak photon flux density is

$$\Phi_{\gamma,\text{peak}} = N_\gamma \frac{4 \ln 2}{\pi d_x d_y} \frac{2\sqrt{\ln 2}}{\sqrt{\pi} \Delta t_X}. \quad (\text{S7})$$

Using $\Delta t_X = 236$ fs, we obtain

$$\Phi_{\gamma,\text{peak}} \simeq 2.1 \times 10^{27} \text{ photons s}^{-1} \text{ cm}^{-2}. \quad (\text{S8})$$

I.C Shot-by-shot extraction of FEL-correlated scattering intensity

For each static data point or pump-probe delay point, data acquisition continued until 1000 valid single-shot CMOS images had been accumulated. A pulse was considered valid only when its GMD2 signal exceeded the preset threshold of 1×10^{-9} A; pulses with insufficient GMD2 intensity were rejected during acquisition and were not counted toward the 1000-shot total. The 1000 valid images were averaged to determine the diffraction-peak position. A fixed circular region of interest (ROI) centered on the peak was then selected, as shown in Fig. S2(a). For each shot j , the raw detector counts inside the ROI were integrated to obtain C_j^{ROI} . The same ROI was used for all shots acquired under the same scattering condition.

Because the FEL pulse intensity fluctuates strongly from shot to shot, the arithmetic mean of the raw ROI counts does not provide a reliable measure of the scattering intensity. We therefore correlated the single-shot ROI counts with the simultaneously recorded GMD2 signal $G_{2,j}$, as shown in Figure S2(b,c). The finite spread of the data points around the overall correlation cannot be attributed solely to pulse-intensity fluctuations. Because the resonant scattering cross-section is sensitive to photon energy, both the energy profile of single

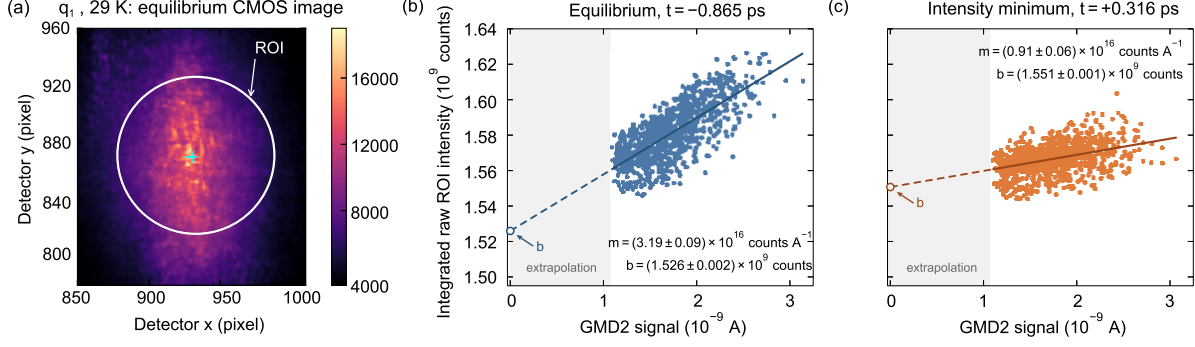


Figure S2: **Shot-by-shot extraction of the FEL-correlated resonant scattering intensity.** (a) Representative averaged CMOS image of the q_1 reflection at 29 K. The white circle marks the region of interest (ROI) used to integrate detector counts. (b,c) Shot-by-shot ROI-integrated detector counts plotted as a function of the simultaneously recorded GMD2 signal for the q_2 reflection in equilibrium (b) and near the pump-induced intensity minimum (c), respectively. Each panel contains 1000 valid FEL shots acquired at a fixed pump-probe delay. Solid lines show linear fits within the measured GMD2 range. Dashed lines in gray shade indicate extrapolation toward $G_{2,j} = 0$. The slope m represents the incident-pulse-normalized FEL-correlated scattering intensity, whereas b_{fix} denotes the ROI baseline independent of the GMD2 signal.

FEL pulses and shot-to-shot jitter of the central photon energy can introduce additional variations in the measured ROI counts that are not captured by the GMD2 signal. The influence of these photon-energy variations/jitters, including their effects on the q_1 and q_2 measurements, is discussed in detail in the following Supplementary Section. For each pump-probe delay t , the shot-by-shot data were fitted using

$$C_j^{\text{ROI}}(t) = m(t)G_{2,j} + b_t, \quad (\text{S9})$$

where $m(t)$ is the fitted slope and b_t is the delay-dependent intercept extrapolated to $G_{2,j} = 0$. The intercept contains additive contributions to the ROI counts, including the detector offset, dark counts, and image background that are not correlated with the recorded GMD2 signal.

Rather than adopting a single fixed intercept, we let $b(t)$ follow the delay-dependent image background, which was scaled from a reference intercept using relative count variations integrated over a diffraction-free reference region.

The slope $m(t)$ quantifies the average increase in ROI counts per unit GMD2 signal and is therefore used as the incident-pulse-normalized scattering intensity. A larger slope means that the same incident FEL pulse intensity produces a stronger resonant scattering signal, whereas a reduced slope indicates suppression of the diffraction peak. All time-resolved intensities reported in the main text [Fig. 1(d) and Fig. 4] were extracted from the fitted slope $m(t)$. The error bar of each intensity point is given by the standard error of the fitted slope and therefore quantifies only the statistical precision obtained from the finite ensemble of nominally 1000 shots at that delay. It does not include systematic deviations of the measured intensity from the intrinsic scattering intensity of the ordered state caused by photon-energy drift.

I.D Photon-energy contributions to the scattered distribution of the shot-by-shot data

Even after accounting for the large pulse-to-pulse variation in FEL intensity using the simultaneously recorded GMD2 signal, the single-shot ROI counts remain distributed over a finite range around the overall ROI-GMD2 correlation. Representative equilibrium distributions measured at 29 K are shown in Fig. S3(a,b). The data for the native q_1 reflection exhibit a substantially broader distribution than those for the XFEL-induced q_2 reflection. GMD2 records the total intensity of each FEL pulse. If variations in pulse intensity were the only source of the shot-to-shot fluctuations, pulses with similar GMD2 signals would produce nearly identical ROI counts and the data points would lie close to a single straight line. Instead, a clear vertical spread is observed: pulses with comparable GMD2 signals can generate substantially different diffraction counts. This shows

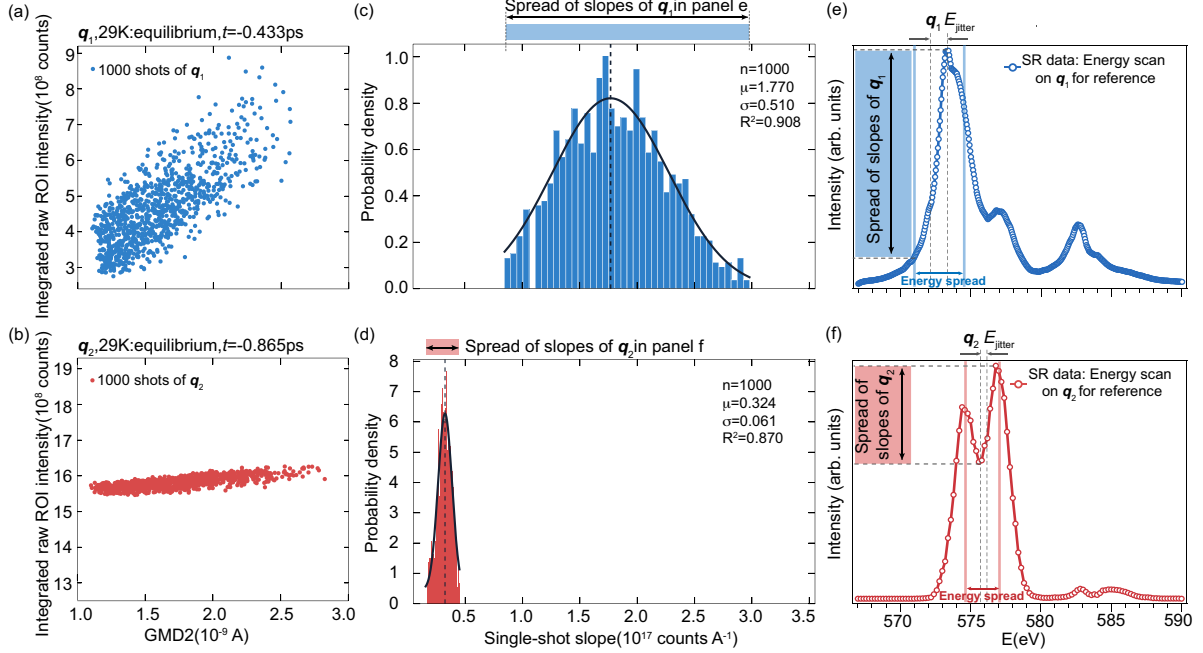


Figure S3: **Photon-energy contributions to the scattered distribution of the shot-by-shot scattering response.** (a, b) Raw ROI counts versus GMD2 for 1000 valid shots measured at 29 K for the equilibrium native q_1 reflection at $t = -0.433$ ps (a) and the equilibrium XFEL-induced q_2 reflection at $t = -0.865$ ps (b). (c, d) Distributions of the single-shot slopes $s_j = [C_j^{\text{ROI}} - b(t)]/G_{2,j}$ obtained from a and b, respectively. Black curves are Gaussian fits, while dashed lines mark their mean values. (e, f) Synchrotron energy scans of the q_1 (e) and q_2 (f) reflections with schematic FEL photon energy profiles with energy jitters.

that the resonant scattering efficiency, namely the detected ROI counts per unit incident pulse intensity, also fluctuates from shot to shot.

To quantify this variation, we calculated an effective response for each valid pulse as

$$s_j(t) = \frac{C_j^{\text{ROI}}(t) - b(t)}{G_{2,j}}, \quad (\text{S10})$$

where $b(t)$ is the intercept determined for the corresponding delay point. The resulting single-shot slope distributions are shown in Fig. S3(c,d). For the representative q_1 data, the distribution has a mean of 1.770×10^{17} counts A^{-1} and a standard deviation of 0.510×10^{17} counts A^{-1} . By comparison, the corresponding q_2 distribution is considerably narrower, with a mean of 0.324×10^{17} counts A^{-1} and a standard deviation of 0.061×10^{17} counts A^{-1} . The relative standard deviations are approximately 29% and 19%, respectively.

Two photon-energy effects can contribute to the scattered distribution of single-shot slopes. First, each FEL pulse has a finite spectral bandwidth of approximately $\Delta E = 1.22$ eV, and its detailed spectral profile can vary from shot to shot. As a result, pulses with similar GMD2 signals may sample different parts of the resonant line shape and produce different diffraction intensities. Second, the central photon energy also exhibits shot-to-shot jitter, which shifts the entire pulse spectrum along the energy axis [Figs. S3(e-f)]. Because the q_1 and q_2 intensities vary strongly with photon energy near the Cr L_3 edge, even a modest energy jitter can result in a higher or lower effective single-shot slope.

I.E Identification and treatment of photon-energy drift

In several pump-probe data sets, we observed an anomalous distribution with a subset of single-shot data forms a relatively isolated low-response branch in the ROI-GMD2 correlation plot, as illustrated in Fig. S4(a)

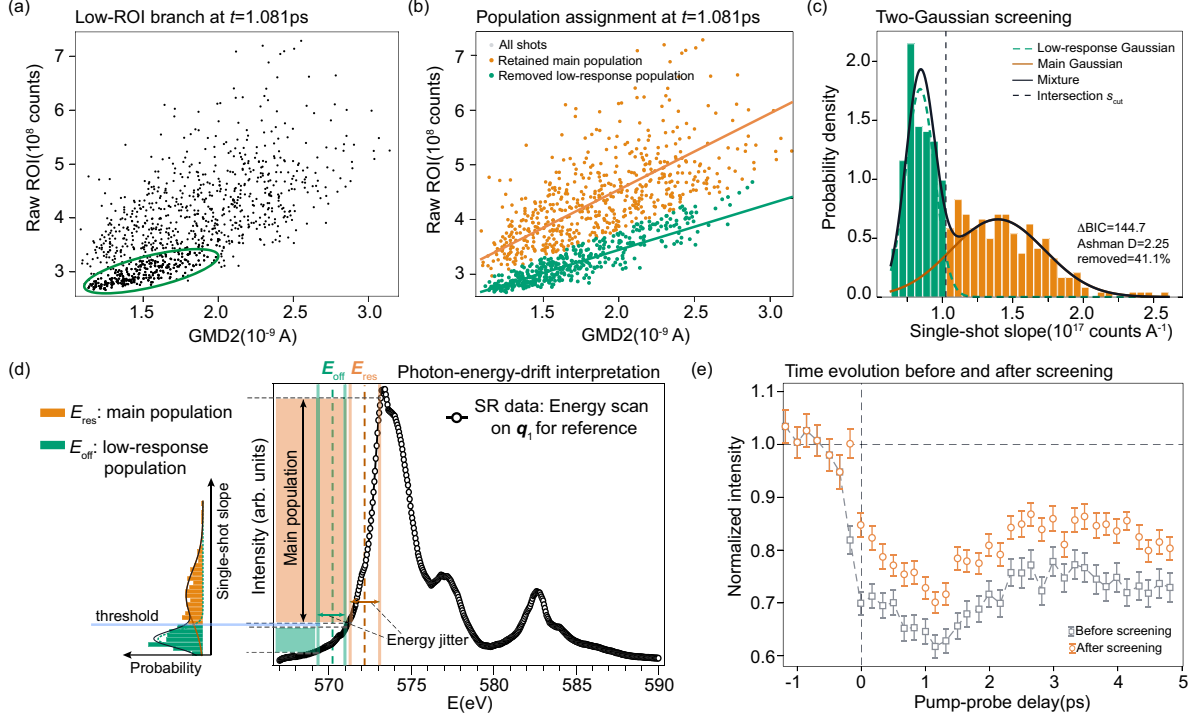


Figure S4: **Identification and treatment of the low-response branch.** (a) Representative shot-by-shot ROI counts versus GMD2 for the q_1 data at $t = 1.081$ ps. A distinct low-response branch is visible at the lower edge of the main data cloud. (b) Same data as panel (a) with the main population and low-response population in different colors. (c) Distribution of the single-shot slopes used to separate the low-response branch from the main resonant population. The threshold was determined from the intersection of two Gaussian components representing the low-response and resonant populations. (d) Schematic illustration of photon-energy drift on the Cr L_3 -edge resonant line shape. A drift of the FEL central photon energy away from resonance produces a reduced scattering response that can form a separate low-response population. (e) Comparison of time-resolved q_1 traces extracted before and after filtering.

for the q_1 data at delay time $t = 1.081$ ps. After subtracting the intercept and normalizing by GMD2, the distribution of the single-shot slope s_j shows an abnormal enhancement at low response values [Fig. S4(b,c)].

The finite FEL spectral bandwidth and ordinary shot-to-shot photon-energy jitter mainly broaden the main data cloud in a single Gaussian profile [Figs. S3(c-d)]. In contrast, the relatively isolated low-response branch in Fig. S4(a-c) suggests that a subset of pulses was displaced farther from the Cr L_3 resonant condition. When the FEL center photon energy drifts below the resonant maximum, the resonant scattering intensity decreases and becomes less sensitive to changes in photon energy, producing a large population of weak scattering response even for shots with comparable GMD2 signals [Fig. S4(d)].

This low-response population was not observed in the negative-delay equilibrium data in Fig. S3(a). Instead, it appeared in part of the positive-delay q_1 pump-probe data as the experiment progressed. This behavior suggests a slow change in the accelerator operating condition, which shifted the center of the photon-energy jitter distribution. We use the term photon-energy drift to describe this experimentally resolvable energy displacement beyond ordinary shot-to-shot jitter.

To identify and reduce the influence of this drift-induced population, we first determined a common intercept b_0 and then constructed a shot-filtering criterion based on the single-shot slope. For each delay point where a low-response population was visible, the distribution of s_j was fitted by two Gaussian components. The lower-response component represents off-resonant or weakly resonant shots, whereas the higher-response component represents the main resonant population. The intersection of the two Gaussian components was used as the filtering threshold. After filtering, only the retained main population was used to refit the ROI-GMD2 correlation with the intercept constrained to b_0 . The resulting slopes were normalized by the

average value of the negative-delay data.

Figure S4(e) compares the time delay traces obtained before and after filtering. The low-response shots make the unfiltered data appear to have a deeper intensity suppression and a lower recovery plateau. Removing these shots yields a trace that more closely represents the resonant response of the $\mathbf{q}_1$ order.

This procedure does not completely eliminate the effect of photon-energy drift. The present experiment did not include a shot-by-shot photon-energy diagnostic synchronized with the CMOS images. We therefore cannot directly determine the central photon energy of each pulse, nor can we fully separate photon-energy variations from the intrinsic sample dynamics. The filtering procedure removes the most prominent low-response population, but smaller continuous energy drifts that do not form a separate branch may remain within the main data cloud and may contribute to systematic intensity variations among delay points.

II Analysis of the synchrotron RSXS data

II.A Extraction of peak position, integrated intensity, and correlation length

Each synchrotron L -scans measured with synchrotron radiation was fitted by a Lorentzian peak on a slowly varying linear background,

$$I(L) = B_0 + B_1L + \frac{A}{1 + [(L - L_0)/\gamma_L]^2}, \quad (\text{S11})$$

where L_0 is the peak position and γ_L is the half width at half maximum (HWHM) in reciprocal-lattice units. The term $B_0 + B_1L$ accounts for the slowly varying baseline within the fitted L range, including possible contributions from the tail of the specular surface reflection and the detector background.

The full width at half maximum is

$$\Delta L_{\text{FWHM}} = 2\gamma_L. \quad (\text{S12})$$

For scans along $(0, 0, L)$, the HWHM in absolute momentum units is

$$\kappa_c = \frac{2\pi}{c}\gamma_L, \quad (\text{S13})$$

and the corresponding observed correlation length is defined as

$$\xi_c = \frac{1}{\kappa_c}. \quad (\text{S14})$$

For the conversion from reciprocal-lattice units to absolute momentum transfer, we used the low-temperature lattice parameter $c = 6.007(9)$ Å, determined at 20 K in Ref. [3]. The same value was used consistently for all temperatures in the present analysis. The temperature dependence of the lattice parameter introduces only a small correction to the absolute momentum scale compared with the observed linewidth variation; therefore, using a fixed low-temperature value does not affect the temperature-dependent trend of the extracted correlation length.

No correction for the instrumental momentum resolution was applied. The reported values of ξ_c were calculated directly from the measured peak widths and therefore represent observed correlation lengths, which provide lower bounds on the intrinsic values. The integrated diffraction intensity was calculated from the fitted Lorentzian area, $I_{\text{int}} = \pi A \gamma_L$. The error bars for the peak position, integrated intensity, and correlation length in the temperature-dependent results of Fig. 2 in the main text represent one-standard-error uncertainties obtained from the Lorentzian fits.

II.B Extraction of resonant-energy profiles

For the energy-momentum maps in Fig. 3 of the main text, the incident photon energy was scanned at each fixed L position. Each energy scan was normalized to the photocurrent of an upstream Au mesh used to monitor the incident photon flux, and the normalized scans were then assembled into the two-dimensional $I(E, L)$ maps.

The resonant-energy profiles of the $\mathbf{q}_1$ and $\mathbf{q}_2$ reflections were obtained by integrating the normalized intensity over the fixed L ranges 0.345–0.367 and 0.383–0.422, respectively. The corresponding integration range was kept unchanged throughout each photon-energy scan.

III Time-resolved RSXS experimental conditions

III.A Optical-pump conditions and temporal resolution

The time-resolved measurements were performed using π -polarized optical pump pulses with a photon energy of 1.55 eV (800 nm) and a duration of 82 fs FWHM. Unless otherwise stated, the pump fluence was 0.57 mJ cm^{-2} . The incident pump beam had an approximately circular Gaussian profile with a FWHM diameter of $203 \mu\text{m}$. Its incidence angle of approximately 32° relative to the sample surface produced an elliptical footprint of approximately $203 \mu\text{m} \times 383 \mu\text{m}$ FWHM, which was used to calculate the fluence.

The π -polarized XFEL probe was tuned to approximately 576 eV, near the Cr L_3 absorption edge, and had a spot size of approximately $57 \mu\text{m}$ FWHM. The probe area was therefore well contained within the optically excited region, ensuring approximately homogeneous excitation over the x-ray-probed area.

The overall pump-probe temporal resolution was primarily limited by shot-to-shot arrival-time jitter between the optical-pump and XFEL-probe pulses. It was experimentally determined from the pump-induced reflectivity change of a YAG reference under the same timing conditions [Fig. S5]. A step response convolved with a Gaussian instrument-response function yielded a temporal resolution of 297 fs FWHM. This experimentally determined value was fixed as the instrument-response width in all dynamical fits described below.

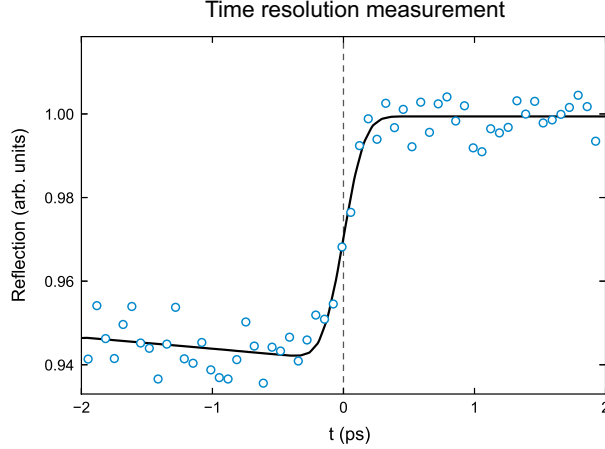


Figure S5: **Experimental determination of the pump-probe temporal resolution.** Transient reflection signal from a YAG reference as a function of pump-probe delay. The open circles represent the measured data, and the solid line is a fit to a step response convolved with a Gaussian instrument-response function. The vertical dashed line marks time zero. The fit yields an overall temporal resolution of 297 fs FWHM, which was used in the analysis of the time-resolved diffraction data.

IV Analysis of the ultrafast dynamics

IV.A Gaussian-convolved two-step recovery model

The time-resolved RSXS traces were analyzed using a two-component recovery model convolved with the experimentally determined Gaussian instrument-response function (IRF), following the framework of Ref. [4]. The two components describe the fast and slow recovery processes following the pump-induced suppression of the diffraction intensity.

The Gaussian instrument-response function is written as

$$G(t) = \frac{1}{\sqrt{2\pi}\sigma_{\text{IRF}}} \exp\left(-\frac{t^2}{2\sigma_{\text{IRF}}^2}\right), \quad (\text{S15})$$

where $\sigma_{\text{IRF}} = 126$ fs corresponds to the experimentally determined temporal resolution of 297 fs FWHM described in Sec. III.A. The IRF width was fixed to this value for all fits and was not treated as a free parameter.

For a two-component recovery, the response is written as

$$I_{\text{int}}(t) = I_0 - H(t - t_0) \left[A_1 \exp\left(-\frac{t - t_0}{\tau_1}\right) + A_2 \right], \quad (\text{S16})$$

where $H(t - t_0) = 0$ is the Heaviside step function. Here, I_0 is the equilibrium diffraction intensity, t_0 is the response onset relative to the experimentally determined pump-probe time zero, and A_1 is the effective amplitudes associated with the fast recovery time τ_1 . The slow recovery process including the extrinsic contribution from the slow drift of the photon energy is approximated by the delay-independent term A_2 .

The measured response is obtained by convolving the intrinsic response with the Gaussian IRF:

$$I_{\text{obs}}(t) = (G * I_{\text{int}})(t) = \int_{-\infty}^{+\infty} G(t - t') I_{\text{int}}(t') dt', \quad (\text{S17})$$

where the asterisk denotes convolution. The finite rise time of the measured suppression is therefore determined by the experimental IRF and does not require an additional intrinsic onset time.

IV.B Dataset-specific application of the recovery model

The two low-temperature time-resolved measurements of the XFEL-induced q_2 order were fitted using the Gaussian-convolved two-component response model in the long-lived limit [Eqs. (S17)]. The Gaussian IRF was fixed to a FWHM of 297 fs. Because the slow component cannot be assigned a reliable finite recovery time in the current delay window, and because it may include contributions from photon-energy drift, A_2 was used phenomenologically to describe a long-lived intensity contribution during the measured time range.

The two q_2 measurements probe the same XFEL-induced order under nominally identical temperature and excitation conditions. We therefore assumed that the fast recovery time τ_1 is an intrinsic timescale of the q_2 state and should be common to both measurements. The run 1 covers a longer delay range and better constrains the long-lived component. We therefore first fitted run 1 to determine τ_1 . The same value of τ_1 was then fixed in the fit to run 2, which also yielded a reasonable description of the data. The difference in

Table S2: **Summary of the dynamical fit parameters.** The Gaussian IRF was fixed to a FWHM of 297 fs. The amplitudes are normalized by the fitted equilibrium baseline $I_{0,\text{fit}}$.

Dataset	Fit range (ps)	$I_{0,\text{fit}}$	A_1/I_0	τ_1 (ps)	A_2/I_0	t_0 (ps)
q_2 , run 1	$-1.0 \sim 5.0$	1.01	0.46	$0.98^{+0.11}_{-0.09}$	-0.029	0.01
q_2 , run 2	$-1.0 \sim 3.2$	1.0	0.36	0.98 (fixed)	0.088	0.02

A_2 between the two runs is attributed primarily to different photon-energy drift conditions rather than to a change in the intrinsic $\mathbf{q}_2$ dynamics. The fitting parameters used for the curves shown in the main text are summarized in Table [S2](#).

Supplementary references

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