

# Enhanced resonant elastic x-ray scattering in XFEL-generated warm dense matter

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## Article

**Keywords:** XFEL, Transient Resonances, Resonant Scattering, Increase in Scattering Cross Section, Nonlinear X-ray Optics, Ultrashort, Ultraintense

**Posted Date:** October 16th, 2026

**DOI:** <https://doi.org/10.21203/rs.3.rs-10750046/v1>

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**Additional Declarations:** There is **NO** Competing Interest.

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# Enhanced resonant elastic x-ray scattering in XFEL-generated warm dense matter

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### Abstract

Irradiation of solids with focused femtosecond (fs) X-ray Free Electron Laser (XFEL) pulses can drive them into Warm Dense Matter (WDM) and populate excited electronic configurations that substantially increase x-ray opacity and the elastic scattering factor by bound-bound resonant transitions. Here, we observe a transient enhancement of Cu elastic scattering near the  $L_3$  edge via  $L_\alpha$  (2p–3d) transitions at 929.7 eV. The excitation sequence starts with photoionization, followed by Auger decay that produces high-energy electrons. These then trigger electron-impact ionization, creating a non-Local Thermodynamic Equilibrium (non-LTE) state. On a fs timescale, Cu ions remain at their cold-solid equilibrium positions while occupying excited electronic configurations with multiple 3d holes. Using XFEL pulses elastically scattered from a Cu-containing multilayer [SiC/Cu/B<sub>4</sub>C], we spectrally resolve the fifth-order superlattice Bragg peak around the  $L_\alpha$  resonance. Its amplitude and spectral width increase nonlinearly with excitation intensity. A self-consistent theory describing the formation of the non-LTE state and its x-ray optical properties links transient electronic configurations to the measured scattering response and reproduces the data. An effective two-level model quantifies a twelvefold enhancement of the imaginary scattering factor relative to cold Cu, demonstrating the role of transient resonances and enabling ionization- and state-selective contrast for ultrafast x-ray imaging.

**Keywords:** XFEL, Transient Resonances, Resonant Scattering, Increase in Scattering Cross Section, Nonlinear X-ray Optics, Ultrashort, Ultraintense

XFELs enable the production of fs and attosecond X-ray pulses with intensities exceeding  $10^{20}$  W cm<sup>-2</sup> [1, 2]. These pulse characteristics enable nonlinear X-ray matter interaction, driving atoms into highly-ionized, non-equilibrium states [3, 4] by sequential inner-shell photoionization and cascaded electronic decays. In addition to photoionization, bound-bound transitions on transient resonances – that are “hidden” in the ground-state by the Pauli exclusion principle and become accessible by creating holes in core or valence electronic orbitals – can play a major role in the creation of highly-charged ions [5–7]. Since the lifetime of a core-excited electronic configuration is at the order of a fs, such resonant transitions are transient. Transient resonances on inner-shell transitions have been studied in early XFEL studies of

nonlinear absorption and resonant Auger effect in simple atomic systems, which were investigated both theoretically [8–10] and experimentally [11, 12]. Furthermore, hidden resonances are key to atomic X-ray lasing [13–15] and stimulated X-ray Raman scattering [16, 17]. Recently, they have been theoretically studied in the context of Resonant X-ray Scattering (RXS) to determine interatomic electron currents [18] and interference in resonance fluorescence signals [10, 19]. Importantly, RXS has been applied as a key element to study plasma formation in nanoparticles [20], exploiting charge-state specific contrast of transient resonances to image the inhomogeneity in the plasma composition.

The efficient photoionization by XFELs at intensities  $> 10^{14} \text{ W cm}^{-2}$  paired with electron collisions drive solids into the regime of WDM that initially is far from local thermodynamic equilibrium. WDM is a state in between plasma and solid and abundant at the surfaces of white dwarfs, interiors of heavy planets and a transient state in inertial-confinement fusion. Theoretical descriptions of the optical properties of WDM and its equation of state are challenging even for thermodynamic equilibrium [21, 22]. The transition of a solid into WDM is accompanied by a stark modification of its x-ray optical properties [23, 24] which are notoriously difficult to theoretically predict. Bound-bound transition condition optical properties [25, 26] and the dynamic structure factor [27, 28]. Mercadier *et al.* [25] recently demonstrated the role of transient resonances in XFEL-generated WDM on the absorption coefficient of warm dense Cu by x-ray absorption at the Cu  $L_3$  and  $L_2$  edges. They identified metastable vacancies in the 3d shell (figure 1c) that open 2p-3d absorption channels below the Cu  $L_3$  edge, effectively increasing the absorption.

Absorption and scattering are intrinsically connected: the absorption coefficient  $\mu$  is proportional to the imaginary part of the atomic scattering factor  $f_2$  via  $\mu = 2r_e\lambda Nf_2$ . Here  $r_e$  is the classical electron radius,  $\lambda$  the wavelength and  $N$  the atomic density. This relation implies that transient resonances must also influence

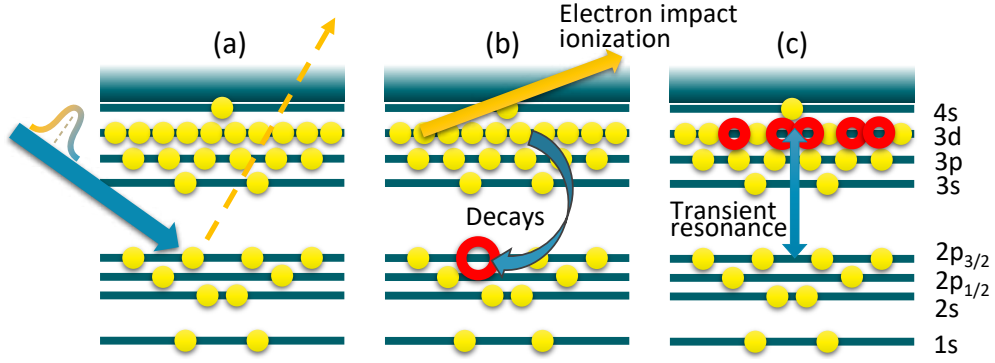


**Fig. 1 Mechanism that leads to the creation of the  $2p_{3/2} - 3d$  transient resonance.** (a) A 2p electron is photoionized by the X-ray pulse. (b) The created hole rapidly transfers to vacancies in the 3d level by Auger decay, creating secondary electrons that thermalize and produce further 3d vacancies by impact ionization. (c) A cascade of electron impact ionization processes establishes a metastable state with abundance of holes in the 3d level, opening the transient 2p-3d resonances.

elastic scattering. One manifestation of this effect is Enhanced Resonant Elastic Scattering (ERES), a process in which the elastic scattering factor of individual atoms is transiently by the additional resonance scattering. An early indication of ERES was reported by Bostedt *et al.* [29], who observed a nonlinear increase in the intensity of xenon nanoparticle scattering patterns with increasing pulse intensity. A more systematic study by Ho *et al.* [30] showed how pulse duration, intensity, and photon energy govern the scattering response of molecules, revealing that transient resonances enhance elemental contrast for short resonant excitation. These observations have been recently corroborated by ultrafast X-ray diffractive imaging [31, 32] for pulse duration below the fs lifetime of inner-shell vacancies. These studies provide evidence of the principle, but they remain limited to spectrally unresolved measurements, which preclude identifying the specific mechanism of the increased scattering.

In this work, we demonstrate spectrally resolved ERES in XFEL generated WDM, exploiting bound-bound transitions in highly-charged Cu ions. We exploit the transiently modified occupancy of the Cu 3d manifold that opens an additional scattering channel (figure 1). By combining spectrally resolved measurements and quantitative predictive modeling, coupling a plasma description of XFEL-driven Cu

excitation [33, 34] with an ab initio multiple scattering calculation to determine optical properties [35], we demonstrate and characterize the enhancement of elastic scattering by transient resonances, providing a basis for applications that benefit from stronger scattering signals or provide state-selectivity by spectrally controlled scattering signals.

## Measurement of spectrally resolved scattering enhancement

To observe the transient modification of the scattering strength of Cu ions upon irradiation with a X-ray pulse we spectrally resolved the radiation diffracted from a Cu-containing multilayer structure – figure 2 shows the experimental setup. The applied self-amplified spontaneous emission (SASE) x-ray pulses featured a highly fluctuating spectrum of an averaged wavelength of 931 eV, a bandwidth (FWHM) of 5 eV (see figure 2b), and a nominative pulse duration of 15 fs. The scattered X-ray spectrum was analyzed by the hRIXS spectrometer [36], integrating the 5-th order superlattice peak over several SASE pulses. Figure 2d shows a typical detector image. The superlattice peak is concentrated in a small area on top of the the Cu- $L_\alpha$  fluorescence spanning over the whole non-dispersive vertical detector axis. See Methods for more details. In our experiment it is a single X-ray pulse that both pumps the cold Cu sample into WDM and serves as a probe pulse. For photon energies above the  $L_3$  edge the x-ray pulse efficiently ionizes Cu, creating a manifold of Cu ions of different charge states and electronic configurations during the course of the pulse. Thus, a plenitude of transient resonances with transition energies below the cold  $L_3$  edge are created within the pulse duration, pertaining to different ionic charge states, electronic configurations and in an evolving plasma environment.

The results of the measurement are presented in figure 3a. The “cold” spectrum, recorded at  $1.2 \times 10^{13} \text{ W cm}^{-2}$ , exhibits the usual anomalous dispersion characteristics

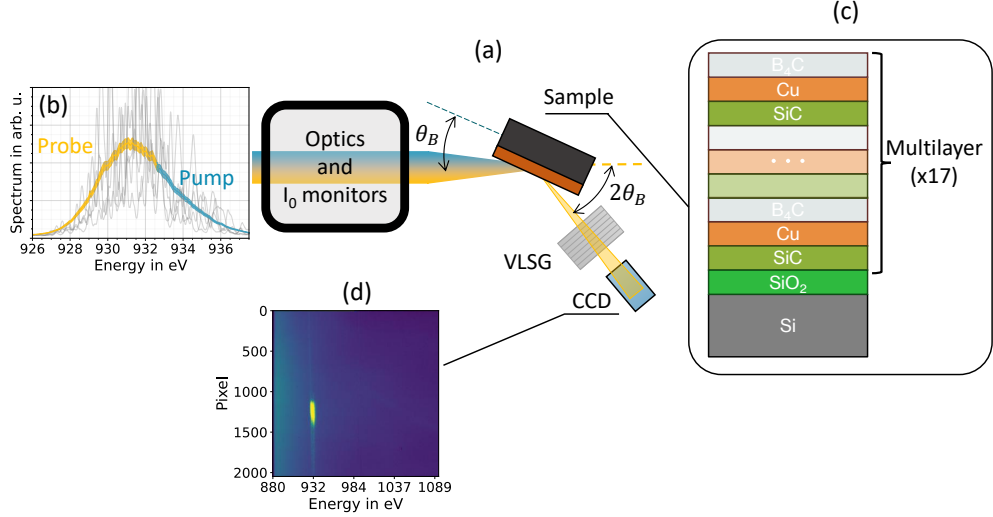


**Fig. 2 Experimental setup.** (a) We use a X-ray pulse with central wavelength 931 eV and bandwidth 5 eV. A single pulse possess both probing components (yellow) below the Cu-L<sub>3</sub> edge (932.7 eV) and pumping components (blue) above the edge. (b) Average spectrum together with a few sample single realizations. The pulse is focused onto the multilayer sample, which is tilted by the Bragg angle  $\theta_B$ . The diffracted radiation is collected into the spectrometer, schematically represented by a grating and the CCD camera. (c) The sample is a nano-structured [SiC/Cu/B<sub>4</sub>C] multilayer, having 17 repetitions, grown on a Si substrate with SiO<sub>2</sub> interface layer. (d) Detector image showing the diffracted signal (bright spot) superimposed with the Cu-L <sub>$\alpha$</sub>  fluorescence line (faint vertical line). The horizontal axis is the dispersive axis of the spectrometer and is mapped to energy, the vertical axis is the non-dispersive direction.

134 near the Cu-L<sub>3</sub> edge at 932.7 eV, composed of the impulsive Thomson scattering and  
 135 the dispersive bound-free scattering contribution for photon energies exceeding the  
 136 L-shell ionization threshold. As the excitation intensity increases, additional scatter-  
 137 ing features emerge in the pre-edge region, most prominently as a peak at the Cu-L <sub>$\alpha$</sub>   
 138 ( $2p_{3/2} - 3d$ ) resonance at 929 eV. This feature constitutes the signature of the tran-  
 139 sient resonance. The resonant contribution grows nonlinearly with intensity, reaching  
 140 a maximum at  $8 \times 10^{14} \text{ W cm}^{-2}$ , where the peak amplitude is nearly six times the base  
 141 value of the cold spectrum. A further increase in intensity reduces the peak amplitude.  
 142 This is caused by the greater number of 3d holes, which decrease the screening of the  
 143 nuclear charge and in turn shift the resonances to higher energies, outside the obser-  
 144 vation window. Concurrently with this trend, the resonant peak spectrally broadens.

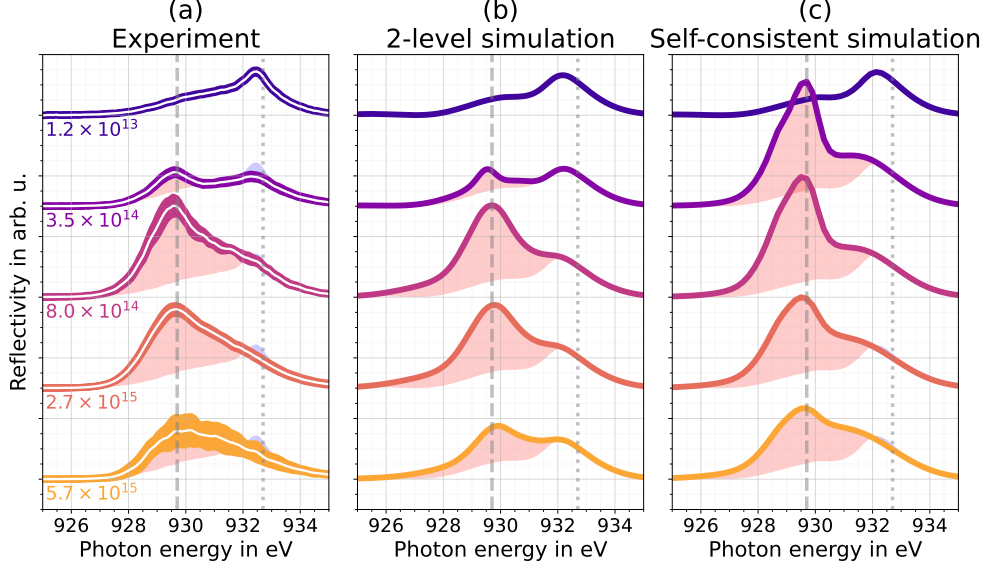


**Fig. 3 Spectrally resolved transient enhancement of the scattering strength of Cu atoms, experimental data, fit with the two-level model and simulation with the self-consistent theory.** (a) Experimental measurement of the spectrally resolved scattered radiation from the multilayer structures around the Cu- $L_{\alpha}$  resonance for different intensities (in  $\text{W cm}^{-2}$ ). The different spectra are offset vertically by -3 arb. u. for clarity. The colored region around each spectrum represents the 99% confidence interval. The shaded red (or blue) regions correspond to positive (or negative) difference with respect to the reference spectrum measured at the low intensity  $1.2 \times 10^{13} \text{ W cm}^{-2}$ . The dashed and dotted lines mark the position of the Cu- $L_{\alpha}$  resonance (929.7 eV) and the Cu- $L_3$  edge (932.7 eV) respectively, at low intensity. (b) Result of the fit of the two-level model to the experimental data. (c) Simulation of the reflectivity curves with the self-consistent model.

145 Importantly, the 5th order superlattice peak is recorded up  $6 \times 10^{15} \text{ W cm}^{-2}$  intensity,  
 146 showing that within the 15 fs measurement time, the structure and interface of the  
 147 multilayer stays intact, with atomic cores resting at their equilibrium position. This is  
 148 corroborated by simulations following the structural evolution of the multilayer upon  
 149 high-intensity x-ray radiation (see SI 2 for details).

## 150 Resonance model for quantification of scattering 151 enhancement

152 Within the independent atom model, the atomic scattering factor close to the Cu L  
153 edge is given by

$$f(\omega) = f_0 + f'(\omega) + if''(\omega), \quad (1)$$

154 with the atomic structure factor  $f_0$  of impulsive elastic scattering of X-rays and the  
155 photon-energy ( $\omega$ ) dependent anomalous scattering contributions  $f'$  and  $f''$  which  
156 give a sizable contribution close to absorption edges and are conditioned by bound-  
157 free scattering processes. Typically, the atomic scattering factor is restructured as  
158 sum of its real ( $f_1 = f_0 + f'$ ) and imaginary ( $f_2 = f''$ ) parts. At room temper-  
159 ature, all values in equation 1 can be taken from tabulated data [37]. For excited  
160 electronic systems, we have to include a bound-bound scattering contribution  $f_T(\omega)$ .  
161 From an atomistic perspective, during the interaction with the x-ray pulse various  
162 resonances appear pertaining to different electronic configurations and charge states  
163 that are transiently occupied in the WDM. For quantifying the resonance scattering  
164 strength and fitting the experimental data, we assume an effective two-level system  
165 characterized by an energy  $\omega_0$  between the initial  $|i\rangle$  and the core-excited state  $|e\rangle$ ,  
166 a spontaneous decay rate  $\Gamma_{sp}$  and an effective width  $\gamma_2$  of the transition. Consid-  
167 ering incoming monochromatic radiation with wavelength  $\lambda$  (photon energy  $\omega$ ) the  
168 additional scattering contribution is :

$$f_T(\omega) = -\underbrace{\frac{3}{8\pi} \frac{\lambda}{r_e} \Gamma_{sp} (\rho_{ee} - \rho_{ii})}_{=A} \frac{(\omega - \omega_0) - i\gamma_2/2}{(\omega - \omega_0)^2 + (\gamma_2/2)^2}, \quad (2)$$

169 where  $\rho_{ee}$  and  $\rho_{ii}$  are the population of the core-excited and initial state. See Sup-  
170 plementary Information (SI) for the derivation. The total atomic scattering factor is

171 then given by:

$$f(\omega) = f_1(\omega) + if_2(\omega) + f_T(\omega). \quad (3)$$

172 We use equation 3 to calculate the  $\omega$ -dependent reflectivity of the multilayer structure  
 173 (see methods and SI for further details), leaving  $\omega_0$ , the amplitude ( $A$ ), and the  
 174 spectral width ( $\gamma_2$ ) as free parameters to be fitted against experimental data. The  
 175 results of this fitting procedure are shown in figure 3b. Figure 4 shows the dependence  
 176 of the parameters of the transient resonance on the pulse intensity. The fit captures  
 177 well the behavior of the transient scattering contribution with the appearance, growth  
 178 and broadening of the resonant peak. The peak value ( $2A/\gamma_2$ ) of the imaginary part of  
 179 the transient atomic scattering factor rapidly grows to its maximum value, roughly 25  
 180 for an intensity of  $8.0 \times 10^{14} \text{ W cm}^{-2}$ , and then drops for higher intensities. The width  
 181 ( $\gamma_2$ ) of the peak has a similar behavior, rapidly growing past the natural width of the  
 182 Cu- $L_\alpha$  emission (0.5 eV) [38], reaching a maximum of almost 4 eV and then steadily  
 183 decreasing. Concurrently, the resonant energy ( $\omega_0$ ) of the effective transition initially  
 184 jumps to lower values, to then gradually increase and shift closer to the Cu- $L_3$  edge  
 185 (932.7 eV).

186 These trends agree with the transient absorption measurements in [25]. With  
 187 increasing intensity and ionization rates, the WDM state is conditioned by a higher  
 188 ionization degree of its Cu ions. With increasing charge-state the resonance shifts to  
 189 higher energies and the distribution of ionic charge states results in broadening of the  
 190 effective resonance. Since  $\omega$  of the incoming pulse is fixed at 931 eV, the transient res-  
 191 onance gradually slips out of the observation window and its intensity drops in the  
 192 reflectivity curve. Extrapolating the trends that we observe in figure 3 towards higher  
 193 intensities ( $>10^{16} \text{ W cm}^{-2}$ ), our plasma model predicts that the peak eventually shifts  
 194 completely above the  $L_3$  edge region, broadens and disappears.

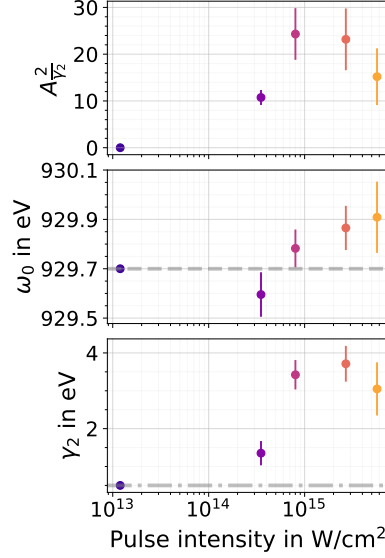


**Fig. 4 Resulting parameters from the fit of the two-level model to the experimental data.** Parameters of the Lorentzian representing the transient contribution to the scattering factor of Cu atoms (see equation 2). The standard deviation as resulting from the fit are multiplied by 5 for better visibility. The parameters for the lowest intensity curve are not fitted, instead they are fixed to the case of zero enhancement ( $A = 0$ ,  $\omega_0 = 929.7$  and  $\gamma_2 = 0.5$ ). The dashed line in the middle panel marks the Cu- $L_{\alpha}$  resonance. The dash-dotted horizontal line in the bottom panel is the width of the Cu- $L_{\alpha}$  emission (0.5 eV) [38].

## Self-consistent simulation of the diffraction spectra

We employed a recently developed quantitative description of the evolution of the non-LTE WDM state [25] that integrates a plasma description of the WDM state along with a solid-state electronic structure calculation capable for calculating the X-ray absorption coefficient and scattering strength. In our framework, a Boltzmann-equation based kinetic approach [33, 34] describes the build up and evolution of the non-LTE plasma from an atomistic perspective, providing a time-dependent abundance of Cu ions of specific charge state and electron configuration, as well as the kinetic-energy distribution of the free electrons. For details on the plasma evolution consult reference [25].

Subsequently, the atomistic results are mapped onto a solid-state description to capture the x-ray absorption near edge structure (XANES) of the WDM state. In

207 order to determine the absorption coefficient at a particular time point, we embed Cu  
 208 ions into an appropriate finite-temperature solid-state environment (with the effective  
 209 temperature being determined by linkage to the Boltzmann description and ranging  
 210 up to  $T_e = 25$  eV), calculate the x-ray absorption coefficient by a finite-temperature  
 211 density-functional theory approach [35] and then weight the coefficients over the  
 212 ion abundances as determined by the Boltzmann code, resulting in an  $\omega$ -resolved  
 213 absorption coefficient  $\mu(x, y, t, \omega)$  for every time ( $t$ ) and space point ( $x, y$  being the  
 214 coordinates within the focal plane). The imaginary part of the x-ray scattering factor  
 215 is then directly related to the absorption coefficient and its real part is obtained via  
 216 the Kramers-Kronig transformation of  $f_2$ . The resulting scattering factor is then used  
 217 in the code to calculate the  $\omega$ -dependent reflectivity curve (see methods). Finally,  
 218 the spectrally resolved reflectivity is computed as the average over all time and space  
 219 points, each weighted by the XFEL field intensity  $I(x, y, t, \omega)$ . The result of this model  
 220 is presented in figure 3c.

221 A general qualitative agreement between theory and experiment is evident, with  
 222 the theoretical framework capturing most spectral characteristics, in particular the  
 223 position and structure of the peaks as well as the formation, broadening, and fading  
 224 of the resonance. Nevertheless, quantitative discrepancies are noticeable, most  
 225 prominently the amplitude of the peak at  $3.5 \times 10^{14} \text{ W cm}^{-2}$  is overestimated. There  
 226 are several uncertainties in the experiment that in the future can be systematically  
 227 addressed: The uncertainty of the intensity is estimated 50%. Furthermore, single-shot  
 228 measurements are unavailable and all data are accumulated over a few tens of shots  
 229 (see the Methods section on data acquisition and analysis). This, together with the  
 230 absence of a shot-by-shot measurement of the fluctuating incident XFEL spectrum,  
 231 limits the accuracy of the spectrally resolved reflectivity – in contrast, the theoretical  
 232 model assumes an idealized Gaussian spectral and intensity profile. The parameter  
 233 with the strongest impact on the spectral profile is the angle  $\theta$  between incident XFEL

beam and the sample surface. In a multilayer mirror,  $\theta$  critically shapes the spectral response: Small angular deviations of  $\theta$  from the optimal Bragg angle alter spectral position and line shape, see for example figure S2 in the SI where a Soft X-ray Reflectivity (SXR) 2D map of angle versus energy is shown. In figure 3c, a single value for  $\theta$  is used for all simulated curves – the one that minimizes the residuals between the measurement and the simulation of the lowest intensity curve – however, because each measurement probes a different region of the sample surface, local strain, layer-thickness variations or slight misalignment of the sample as it is rastered may alter  $\theta$  for every single-shot exposure. To highlight the sensitivity on  $\theta$ , in the SI (figure S6) a comparison between the experimental data and the self-consistent simulation is presented, where  $\theta$  differs by 0.05 deg from the one used here. All these limitations can be in principle overcome by designing dedicated beam-lines and measurement schemes.

## Summary and future perspectives

We have measured the spectrally resolved enhancement of elastic scattering in warm dense Cu on transient bound-bound  $2p_{3/2} - 3d$  resonances in non-LTE conditions generated under intense XFEL irradiation. Based on an effective two-level model integrated within a simulation of the multilayer reflectivities, we extracted a transient response of  $f_2 \approx 25$  from the experimental data, to be compared with the equilibrium value of  $f_2 \approx 2$ . Beyond providing the first spectrally resolved verification of ERES on transient resonances, these results deliver a route to extract transient atomic scattering factors from diffraction spectra and to validate them against microscopic theories of the WDM electronic structure. The next envisioned experiment is a two-color pump-probe setting with probe pulses of sub-fs duration and controllable delay, measuring the transition from non-LTE conditions to equilibrium and supporting benchmark studies of different theories of WDM. With the detailed information on ionization and configuration-state specific optical constants of WDM, ERES could

260 then be extended to the hard x-ray range and for nanometer-scale imaging of hot  
 261 solid-density plasma [39–41]. Along similar lines, ERES could find applications in  
 262 state-resolved imaging of spatial heterogeneity in phase transitions [42]. Another field  
 263 that could greatly benefit from controllable enhancements of the scattering properties  
 264 of atoms is Single-Particle Imaging (SPI) – X-ray diffraction for structure determina-  
 265 tion on non-crystalline objects such as single biomolecules or nanoparticles. In SPI,  
 266 even with high-repetition rate XFELs and advanced data analysis techniques [43]  
 267 atomic resolution is still out of reach due to weak diffraction signals [44]. In addition  
 268 to the enhanced scattering strength, ERES could also serve for phasing diffraction  
 269 patterns, in the realm of multi-wavelength anomalous diffraction. The transient scat-  
 270 tering enhancement mechanism is likewise expected to apply in the hard X-ray regime,  
 271 targeting K-edges, provided the availability of sub-fs to attosecond pulses needed to  
 272 match the short lifetime of the relevant transient resonances. With these settings  
 273 and leveraging two-color control, the effective enhancement could yield values of  $f_2$   
 274 reaching the hundreds under favorable conditions. Such operation would open routes  
 275 to resonance-assisted innovative methods in SPI, and to state-selective scattering  
 276 schemes that exploit transient increases in  $f_2$  for contrast, resolution and phasing of  
 277 the diffracted intensities.

## 278 **Methods**

### 279 **Experimental setup**

280 The experiment was performed at the Spectroscopy and Coherent Scattering (SCS)  
 281 instrument of the European X-ray Free Electron Laser (EuXFEL) facility. A detailed  
 282 overview of the instrument is reported in [45]. Upon generation in the SASE3 undu-  
 283 lator tunnel, the pulses propagate through a X-ray Gas Monitor (XGM). This device  
 284 measured the total combined pulse energy of the pulses [46]. Following this, the pulses  
 285 passed through a Gas ATTenuator (GATT), enabling a pulse-energy reduction on the

level of a few percent [47]. In the SCS experimental hutch, the combined energy of the pulses was measured by another XGM. Then, the pump and probe beams were focused down to 20  $\mu\text{m}$  radius (at  $1/e^2$ ) by the Kirkpatrick-Baez (KB) optics onto the sample surface which forms an angle of  $\theta$  with the incoming beam direction (close to the Bragg angle  $\theta_B$  of the 5th superlattice peak). The scattered radiation propagates in the spectrometer arm which is set to the corresponding  $2\theta$  angle. The sample itself was mounted, manually glued to a sample holder, inside the X-ray Resonant Diffraction (XRD) chamber, on a stage which allowed for linear movement in all 3 directions plus rotation around the vertical axis. The scattered probe pulse was collected into the Heisenberg-RIXS (hRIXS) spectrometer [36], which was positioned at the appropriate  $2\theta_B$  angle. The radiation was dispersed by a Variable Line Spacing Grating (VLSG) with 1000 lines  $\text{mm}^{-1}$ , resulting in a resolution of 0.27 eV. The spectrum was then recorded by a CCD camera. The energy axis was calibrated using the beamline monochromator [48], and the monochromator calibration was previously checked by a X-ray Emission Spectroscopy (XES) measurement at the Cu-L<sub>3</sub> edge.

## Samples

The samples are nano-structured multilayers mirrors on a Si substrate. The multilayer is composed of 17 repetitions of the tri-layer [SiC/Cu/B<sub>4</sub>C] and designed in such a way, that it has a superlattice diffraction peak for a photon energy of 929 eV and in accordance with the geometrically accessible angles of the hRIXS spectrometer setup. By growing every individual layer with a thickness of 2 nm the fifth order super-lattice peak falls at the angle of  $\theta_B = 34^\circ$ . In figure S7 of the SI, the left panel shows a Scanning Electron Microscope (SEM) picture of the lateral section of the sample before irradiation. The difference between the Cu and the other layers is clearly visible, however the SiC and B<sub>4</sub>C layers are indistinguishable due to similar density. The right panel presents an optical microscope picture of the surface of the sample after

312 irradiation. The 3 vertical series of craters are left by pulses at different intensities.  
313 The terraces at the right side of the main crater are individual layer groups that  
314 are ablated. The same type of damage was previously observed in multilayers under  
315 similar conditions [49].

## 316 **Measurement mode**

317 Achieving a high photon flux ( $>10^{12} \text{ W cm}^{-2}$ ) was essential to reach the nonlinear  
318 interaction regime. However, this condition limited each sample exposure to a single  
319 destructive shot. Consequently, the irradiated spot on the sample had to be moved  
320 aside, and a fresh area needed to be brought into the interaction region with each shot.  
321 The XFEL repetition rate was set to a single pulse per train, allowing the stepper  
322 motor to renew the sample at a rate of 10 Hz. The motor did not step from one point to  
323 the next but moved continuously within user-defined upper and lower sample limits,  
324 which could range from a few mm to a maximum of 2 cm. The speed of the motor  
325 was set to  $0.5 \text{ mm s}^{-1}$ , granting a distance between consecutive shots of  $50 \mu\text{m}$ , this  
326 allowed to irradiate a fresh spot at every XFEL pulse. A post-exposure image of the  
327 sample surface, right panel of figure S7 in the SI, confirms that the motor settings were  
328 adequate. The CCD camera (Princeton Instruments, Model PI-MT3) had a readout  
329 time of 4 s which was incompatible with 10 Hz operation. Therefore, we used a long  
330 exposure time matched to the duration of a full line scan. In this way, each line on  
331 the sample provided a single spectrum (averaged over the number of shots per line),  
332 and from one to an arbitrary number of lines could be measured during one run.

## 333 **Simulation of the reflectivity spectrum**

334 The reflectivity spectra were simulated using a recursive Parratt formalism [50], imple-  
335 mented in an in-house code to allow direct control over the atomic scattering factors.  
336 The structural parameters of the multilayer were obtained from hard X-ray reflec-  
337 tivity measurements and subsequently used as input for the soft X-ray reflectivity

338 calculation. The forward model was benchmarked against high-resolution reflectivity  
 339 measurements performed at the PTB beamline of the BESSY II synchrotron, con-  
 340 firming that it accurately reproduces the spectral position and width of the measured  
 341 feature. Starting from this validated Parratt framework, two related models were used  
 342 to describe the transient response. In the first, referred to as the two-level model,  
 343 the transient contribution to the Cu scattering factor is the one in equation 2.  $A$ ,  
 344  $\omega_0$  and  $\gamma_2$  are free parameters to be fitted against experimental data. In the second,  
 345 called self-consistent model, the scattering factor was directly taken from the self-  
 346 consistent electronic-structure model described in the main text. Further details on  
 347 the simulation are collected in the SI.

## 348 Data analysis

349 Each run is first individually checked; this procedure discriminates between the usable  
 350 and corrupted data. Corrupted data are for example accidental darks, non-stationary  
 351 measurements, measurements on spoiled parts of the sample and any user-induced  
 352 error. Subsequently, measurement series are grouped together to build the final result.  
 353 All steps of data ingestion and preprocessing utilize the SCS toolbox python module  
 354 [51], while the main part of the data analysis is achieved with user written python  
 355 scripts and jupyter notebooks.

356 The initial screening of individual runs consists of the following steps. Some broken  
 357 lines along the non-dispersive direction of the detector are corrected by assigning to  
 358 them the average value of neighboring lines. The detector image is then integrated to  
 359 obtain a 1D spectrum and a polynomial background is removed. All spectra in which  
 360 the integrated value is below a threshold are discarded, this step is needed to remove  
 361 accidental dark images and low quality data coming from the sample edges. The  
 362 total exposed pulse energy, which is the sum of the intensity of every incoming pulse  
 363 composing a line scan, is calculated and used to normalize each individual spectrum.

364 During this final step, images with a large variation of total exposed energy are also  
365 removed. Finally, runs measured at identical conditions are grouped together to obtain  
366 average spectra and confidence intervals.

## 367 Data and codes availability

368 Data recorded for the experiment at the European XFEL are available at [52]. The  
369 codes used for data analysis are available at [53]. The codes and data for the simulation  
370 of the transient reflectivity curve that support the findings of this study are open-  
371 source at [54]. The code XTANT-3 is publicly available from [55].

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537 **Declarations.** The authors declare no competing interests.

538 Ethics declaration: not applicable.

539

540 **Acknowledgements.** We acknowledge European XFEL in Schenefeld, Germany,  
541 for provision of X-ray free-electron laser beamtime at SCS SASE3 under proposal num-  
542 ber 3154 and would like to thank the staff for their assistance. D.R. is part of the Max  
543 Planck School of Photonics supported by the German Federal Ministry of Research,  
544 Technology and Space (BMFTR) and the Max Planck Society. We thank Cammille  
545 Carinan for support with the data analysis. We thank Sabrina Schneider and Harumi  
546 Nakatsutsumi for technical help with sample preparation and characterization. This  
547 work was further supported by the Cluster of Excellence “CUI: Advanced Imaging  
548 of Matter” of the Deutsche Forschungsgemeinschaft (DFG)—EXC 2056—project ID  
549 390715994. Computational resources for XTANT-3 calculations were provided by the  
550 e-INFRA CZ project (ID:90254), supported by the Ministry of Education, Youth and  
551 Sports of the Czech Republic. N.M. thanks the financial support from the Czech Min-  
552 istry of Education, Youth, and Sports (grant nr. LM2023068), and from the European

553 Commission Horizon MSCA-SE Project MAMBA [HORIZON-MSCA-SE-2022 GAN  
554 101131245]. B.Z. acknowledges the funding received from the Collaboration Grant of  
555 the European XFEL and the Institute of Nuclear Physics, Polish Academy of Sci-  
556 ences. J.J.K. and J.J.R. are supported by TIMES at SLAC funded by which is DOE  
557 BES Contract DE-AC02-76SF0051.

558 **Authors' contributions.** D.R., A.B., L.M. and N.R. conceived the experiment.  
559 D.R., A.B., L.M., E.Z., T.F., F.K., C.B., D.K., A.L., N.G., B.v.K., G.M., S.P., J.S.,  
560 M.T. and N.R. performed the experiment at the SCS instrument of EuXFEL. S.S.  
561 tuned the accelerator. D.R., L.M., and T.F. analysed the data. D.R., A.B., M.S.,  
562 B.Z., J.J.K. and J.J.R. performed the theoretical calculations. M.P. characterized the  
563 multilayer samples. S.B. developed the multilayer systems and samples. E.B. prepared  
564 backup samples. C.L. and M.K. performed the soft X-ray reflectivity measurements.  
565 D.R., A.B., L.M., N.R., S.B. and H.C. interpreted the data. D.R. wrote the manuscript  
566 with input from all authors.

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