Mapping ultrafast ionization of atoms and clusters with terahertz-streaking delay

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We apply THz-field streaking to temporally resolve the ultrafast ionization of neutral cluster vs atomic targets exposed to intense ultrashort soft x-ray pulses from a free-electron laser (FEL). In the experiment pristine Xe and mixed Xe/Ar clusters as well as Xe atoms are ionized in the vicinity of the $4d \rightarrow \epsilon f$ giant resonance of Xe. We compare the relative streaking delay between the center-of-mass oscillations of electrons that have final kinetic energies in the spectral region of the Xe($4d$) photoline. Our results show that clusters are ionized at the beginning of the 100 fs FEL pulse as supported by calculations of target frustration in the focal volume. We have identified a significantly larger 40 fs relative streaking delay between mixed Xe-Ar clusters vs Xe atoms compared to a 15 fs relative delay found for pristine Xe clusters. This is attributed to the high sensitivity of our spectroscopic measurement to the degree of condensation of the cluster target. Our results show that THz streaking is a powerful technique to temporally study electron emission from extended targets under intense FEL radiation on time scales that are significantly shorter than the FEL pulse duration.

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I. INTRODUCTION

The remarkable development of short-wavelength free-electron lasers (FELs) as well as laboratory-scale sources based on high harmonic generation have opened up a new field of ultrafast x-ray physics with unique opportunities for the studies of structure and dynamics of matter on their natural time and length scales [1–3]. High irradiation intensity combined with short pulse duration provided access to a new parameter regime of nonlinear x-ray physics [4–7] and has led to the observation of novel phenomena such as x-ray induced transparency [8], collective plasmlike states [9], or interatomic Coulombic decay cascades [10]. Ultrafast time-resolved spectroscopy using FEL pulses is now able to trace the dynamics of electronic and nuclear rearrangement in molecules [11,12]. Another prominent example is the serial time-resolved crystallography that has provided unprecedented insights into the conformational changes triggered by the excitation of Photosystem II with visible light [13].

For many applications, the understanding of the electron dynamics of a system exposed to ultrashort and ultraintense FEL pulses is essential. When a nanoscale system is hit by an intense FEL pulse, complex electron and nuclear dynamics evolve on time scales from a few femtoseconds to several hundreds of picoseconds [14–16]. While the processes evolving in the nanoscale system after the FEL pulse has

ended are accessible in the time domain by one- or two-color pump-probe techniques [17–22], time-resolved studies covering time scales shorter than the FEL pulse duration remain challenging [23]. A technique capable of accessing these time scales is light-field streaking that is sensitive to the time instance at which an electron is freed into the continuum [24–26]. This sensitivity allows one to resolve the electron dynamics within the XUV pulse [24,27,28]. Light-field streaking was applied to map the time difference between electrons emitted from two different orbitals of the same atom [27] or from localized vs delocalized states of a solid sample [28] with attosecond time resolution. Recently this technique has been applied to study electron scattering in dielectric nanoparticles [29]. In the current study we use the streaking delay [27–29] to resolve how fast neutral clusters become ionized by an intense FEL pulse.

II. EXPERIMENT

The principle of our experimental approach is presented in Fig. 1. Figures 1(a) and 1(b) (contour plot) show the electron kinetic-energy distributions vs electron birth time, as calculated for Ar₁₄₇ clusters using a quasiclassical molecular dynamics approach [30]. Three distinct signatures in the electron kinetic-energy spectrum have been studied previously [31,32]. (i) The spectral region around the atomic photoline corresponds to electron emission from neutral clusters [blue shaded area in Figs. 1(a) and 1(b) marks the Ar($3p$) photoline]. (ii) A plateau region that is formed of electrons emitted from

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FIG. 1. Principle of the THz streaking experiment. (a),(b) Results of molecular dynamics simulations (reproduced from Ref. [30]) showing electron kinetic-energy distribution vs electron birth time (contour plot) and electron spectrum (left panel) for two different XUV intensities. The simulation was performed for Ar_{147} clusters exposed to a 30 fs FWHM FEL pulse (violet dashed line) of 38 eV photon energy. (c),(d) Streaking spectrograms simulated for corresponding time-energy distributions.

already ionized clusters. These electrons are energetically downshifted due to the growing electrostatic potential of the cluster. (iii) An energy tail with maximum yield at zero kinetic energy. This part of the spectrum is formed by electrons that are initially trapped by the cluster potential and that are emitted from the expanding cluster at a later stage. Note the shift of the electron emission from the spectral region of the Ar(3p) line (the blue shaded area) towards early time with respect to the FEL pulse maximum. For higher irradiation intensity the shift becomes larger [compare Figs. 1(a) and 1(b)]. According to our calculations of corresponding streaking spectrograms [25,26] the shift can be accessed in the time domain [Figs. 1(c) and 1(d)]:

$$p_{\text{final}}(\tau, t) = p_{\text{in}}(t) - eA_{\text{max}} \cos[\omega(t - \tau) - \varphi_0], \quad (1)$$

where p_{in} and p_{final} are the initial and final distributions of the electron momenta (we consider electrons that are emitted along the polarization direction of the laser dressing field), e is the electron charge, A_{max} is the amplitude of the vector potential, ω is the angular frequency of the dressing field, t describes the time axis of the ionizing pulse (with the time zero set to the pulse maximum), τ is the relative time delay between the ionizing pulse and the dressing field, and φ_0 is the initial phase of the dressing field. Since the dynamics takes place on a femtosecond time scale, a dressing field in the terahertz (THz) frequency region is used [33,34]. In the calculation we use $A_{\text{max}} = 4 \times 10^{-7} \text{ V s/m}$ corresponding to a maximal electric-field amplitude of 6 MV/m, $\omega = 2\pi/T$ with an oscillation period of $T = 400 \text{ fs}$, and $\varphi_0 = \pi$.

The Ar(3p) line in the calculated streaking spectrograms [Figs. 1(c) and 1(d)] shows a characteristic oscillatory behavior that follows the oscillation of the vector potential $A_{\text{THz}} = A_{\text{max}} \cos[\omega(t - \tau) - \varphi_0]$ (blue dashed line). In this plot A_{THz}

is scaled to the kinetic-energy shift ΔW according to

$$\Delta W_{\parallel} \approx -\sqrt{\frac{2W_0}{m_e}} eA_{\text{THz}}, \quad (2)$$

where m_e is the electron mass and W_0 is the kinetic energy of the electron without the laser dressing field. In streaking spectrograms the shift of Ar(3p) emission from neutral clusters towards the leading edge of the FEL pulse appears as a shift of the Ar(3p) center-of-mass (COM) oscillation with respect to the oscillation of $-eA_{\text{THz}}$ [Figs. 1(c) and 1(d), solid and dashed lines].

In order to resolve the ionization of neutral clusters vs atoms in our experiment, we chose pristine Xe and mixed Xe/Ar clusters. The FEL photon energy was in the vicinity of the $4d \rightarrow \varepsilon f$ giant resonance of Xe [35]. We note that the experimental parameters used in our study differ from parameters used in the simulation (Fig. 1). This, however, does not change the physical mechanism that is responsible for the ionization of clusters [36]. The experiment was performed at BL 3 of the Free-Electron Laser facility in Hamburg (FLASH), which provides intrinsically synchronized XUV and multicycle THz pulses [33,37]. At the experimental end station the XUV FEL pulses at 13.5 nm wavelength (92 eV photon energy) were colinearly overlapped with THz pulses of 2.5 THz central frequency (120 μm wavelength). A Mo/Si multilayer mirror (focal length $f = 2100 \text{ mm}$) was used to adjust the XUV and THz beamline lengths and to focus the XUV beam. The XUV peak irradiation intensity in the interaction region was about $9 \times 10^{13} \text{ W/cm}^2$ by assuming a focal spot size of about 30 μm full width at half maximum (FWHM), 100 μJ average FEL pulse energy, and 100 fs (FWHM) pulse duration (estimated from parameters of the FEL operation). An off-axis parabolic mirror ($f = 320 \text{ mm}$) was employed to focus the THz beam, leading to a focused spot size of 1.8 mm (FWHM). The maximal THz electric-field strength was 6 MV/m as estimated from streaking spectrograms. With an estimated Rayleigh length of 30 mm and a target length of about 1.5 mm the estimated Gouy phase shift [38] of the THz field that introduces a change of the XUV-THz delay over the interaction region was about 5 fs. A delay stage available in the THz brunch was used to vary the relative time delay of the THz pulses with respect to the XUV pulses in steps of 30 fs. A time-of-flight spectrometer [33,39] was used to record photoelectron spectra parallel to the polarization of the THz field. Electron spectra were averaged over about 100 single FEL shots at each THz-XUV time delay and arranged into streaking spectrograms.

Clusters were produced at room temperature by an adiabatic expansion of pure Xe gas or a gas premix of 2% Xe in Ar through a pulsed 100 μm conically shaped nozzle with a half-opening angle of 15° . The cluster jet was sent through two skimmer systems to reduce the contribution of uncondensed gas. The pressure within the experimental chamber was maintained at $5 \times 10^{-7} \text{ mbar}$. Additionally a piezodriven slit was installed close to the interaction region to further restrict the cluster beam. The stagnation pressure behind the nozzle was varied between 6 and 22.7 bar in order to produce Xe clusters with an average number of atoms $\langle N \rangle$ between 5700 and 130000 as estimated by scaling laws [40]. Expansion of

FIG. 2. THz streaking spectrograms (left panels) and corresponding THz-field free-electron kinetic-energy spectra (right panels) recorded in atomic (a),(b), pristine Xe cluster (c),(d), and mixed Xe/Ar (e),(f) cluster targets. The dash-dotted lines in streaking spectrograms correspond to the center-of-mass approximations of the Xe(4d) spectral region.

the Xe-Ar gas premix at 17 bar stagnation pressure resulted in the generation of mixed Xe/Ar clusters with an average number of atoms $\langle N \rangle \approx 4000$ [41,42]. According to previous investigations this results in mixed Xe/Ar clusters with about 800 Xe atoms preferably located in the core and a few layers of Ar atoms on top of it [41–43].

III. RESULTS AND DISCUSSION

Figure 2 shows streaking spectrograms (left panels) and the corresponding THz-field free-electron kinetic-energy spectra (right panels) measured upon ionization of Xe atoms (a),(b), pristine Xe clusters (c),(d), and Xe/Ar clusters (e),(f). All spectrograms were measured within the same delay range. Due to the frequency chirp of the FEL pulse [33] the spectrum is broader on the rising slope of the vector potential $-eA_{THz}$ and narrower on the falling slope. Additional changes of the signal level might be caused by changes of the average FEL pulse energy during the scan time (11 min). The cluster electron spectra are similar to the simulated spectrum in Fig. 1(a) and show the spectral contributions of the atomic photoline [Xe(4d) electrons] and a plateau region as well as a contribution from a nanoplasma with a maximum yield at zero kinetic energy [30–32,44,45]. In Xe/Ar clusters the signature at ~ 13.7 eV [Fig. 2(f)] is due to correlated electronic decay taking place during the cluster expansion [39]. In order to increase the visibility of the Xe(4d) spectral region the nanoplasma contribution was fitted with an exponential function [red dashed line in (d) and (f)] and subtracted [green line in (d) and (f)] in the cluster spectrograms. The dash-dotted lines (Fig. 2, left panels) map the COM oscillation

FIG. 3. Center-of-mass values (circles) and the corresponding approximations (solid lines) of the Xe(4d) spectral region for atoms vs pristine Xe clusters (a) and for atoms vs Xe/Ar clusters (b). Data are extracted from the corresponding THz-streaking spectrograms of Fig. 2.

of the Xe(4d) spectral region. The following procedure has been used to approximate the COM values. (i) The region of interest is selected between 21 and 26 eV to cover the oscillation of the Xe(4d) spectral region. At each time delay of a streaking spectrogram the COM value is calculated within a defined region of interest (21–26 eV) in the electron spectrum. Figure 3 shows COM values (circles) calculated from corresponding atomic and cluster streaking spectrograms of Fig. 2 (left panels). (ii) The COM data are approximated by Eqs. (1) and (2) to describe the THz-XUV time-delay dependent kinetic-energy shift ΔW (solid lines in Fig. 3). To find the phase of the cosine term in Eq. (1) we perform a Fourier analysis of COM data and extract the phase of the Fourier component at the central frequency of the THz streaking field. The estimated uncertainty in the approximation of the COM data with the cosine function is ~ 6 fs.

A relative shift of the COM oscillation from clusters vs atomic targets towards early THz-XUV time delays is visible in Fig. 3. This demonstrates a faster ionization of a neutral cluster target with respect to atoms as expected from calculations in Fig. 1. Notably, within the studied cluster size range the average shift of the COM Xe(4d) oscillation between Xe atoms and Xe clusters was the same, namely, 14 fs for $\langle N \rangle \sim 5700$ and 11 fs for $\langle N \rangle \sim 56000$, as well as 14 fs and 16 fs obtained in two consecutive measurements on $\langle N \rangle \sim 130000$ clusters. From two consecutive measurements on mixed Xe/Ar clusters composed of $\langle N \rangle_{Xe} \sim 800$ and $\langle N \rangle_{Ar} \sim 3200$ the extracted COM shift was larger, namely, 40 fs and 38 fs, respectively.

We explain the shorter streaking delay found in pristine Xe clusters by the presence of the uncondensed Xe gas in the cluster target. Indeed, the gas mixture used to produce Xe/Ar clusters consists of only 2% Xe. According to previous investigations the Xe enrichment factor in mixed clusters is ~ 10 [41–43]. By the coexpansion of the Xe/Ar gas premix Xe atoms nucleate in the core. Ar atoms act as a seeding gas and partially evaporate off from the surface of a growing Xe cluster [41,43]. Therefore, the contribution of Xe(4d) electrons from uncondensed gas is substantially eliminated in the Xe/Ar cluster target. This finding is attributed to the high sensitivity of the spectroscopic measurements to the degree of the target condensation.

To understand the insensitivity of the streaking delay to the cluster size we have modeled the ionization of neutral targets

FIG. 4. Model calculations of the ionization of neutral targets in the focal volume. Calculated time-energy distributions of the electron emission for atomic (a) and cluster (b) targets. COM_T marks the center-of-mass coordinate of the temporal emission profile. It is calculated on the time axis of the FEL pulse as a reference frame. Streaking spectrograms calculated for corresponding atomic (c) and cluster (d) time-energy distributions. (e),(f) The center-of-mass coordinate COM_T calculated for the target with increasing number of atoms. Cluster sizes investigated in the current study are marked by dashed lines.

in the focal volume. Ionization was calculated numerically by considering a one-photon ionization rate:

$$\Gamma = N_{\text{atoms}} \sigma \Phi, \quad (3)$$

where N_{atoms} is the number of atoms in the system, $\sigma = 25$ Mbarn is the absorption cross section of Xe [35], and $\Phi = I/\hbar\omega$ is the photon flux density. In the interaction region we have assumed a Gaussian radial and temporal intensity distribution with the peak intensity of $I_0 = 9 \times 10^{13} \text{ W/cm}^2$. The sampling interval of the pulse pedestal has been restricted to ± 130 fs on the time axis and to $75 \mu\text{m}$ for the radial profile. The intensity dependence along the FEL beam propagation axis was neglected due to the long (~ 2.5 mm) Rayleigh length. Figure 4 shows calculated time-energy distribution for the electron emission from atoms (a) and clusters (b) with $\langle N \rangle = 56000$. We have assumed a Gaussian profile for the electron kinetic-energy distribution [vertical axis in Figs. 4(a) and 4(b)]. In contrast to the full molecular dynamics simulation shown in Figs. 1(a) and 1(b) only the emission of a first electron from neutral clusters has been considered in Fig. 4(b). Our calculations show a strong ionization of atomic target already in the rising slope of the FEL pulse. Note that the one-photon ionization rate of clusters [Eq. (3)] is substantially higher due to the large number of atoms N_{atoms} . This results in a much faster ionization of clusters compared to atoms.

The electron streaking [Figs. 4(c) and 4(d)] tracks the coordinate of the COM of the electron emission on the time

axis of the FEL pulse. We refer to it as COM_T (where T stands for the FEL time axis) in order to distinguish it from the COM shift of the Xe(4d) photoline oscillation we have referred to so far. In the experiment the time axis of the FEL pulse is unknown; therefore, only the relative difference between COM_T coordinates in clusters vs atoms can be compared. We have identified $\text{COM}_T = 75$ fs for atoms and $\text{COM}_T = 130$ fs for clusters of $\langle N \rangle = 56000$, resulting in a 55 fs relative streaking delay between atoms and clusters. This result is within the reasonable agreement with the 40 fs extracted in the experiment for Xe/Ar mixed clusters. Deviations can be explained mainly by shot-to-shot fluctuations of the temporal and spatial intensity distributions of FEL pulses and, additionally, by the presence of residual uncondensed gas as well as by electrons emitted from ionized clusters that have kinetic energies between 21 and 26 eV. Figures 4(e) and 4(f) show the COM_T coordinate calculated as a function of the number of atoms in the system. Note that the COM_T increases very fast [Fig. 4(e)] and reaches a saturation value for $N \sim 500$ [Fig. 4(f)]. Due to the fast ionization of clusters with $N > 500$ the streaking delay is insensitive to the studied cluster size [see Fig. 4(f), dashed lines].

IV. SUMMARY

We have exploited the sensitivity of the light field streaking technique to the instance of the electron emission to resolve how fast neutral clusters with respect to atoms become ionized under intense FEL radiation. The ultrafast ionization of a target in the focus volume is crucial for the understanding of multiphoton and nonlinear x-ray physics phenomena [4–10]. The delay in streaking spectrograms reveals ultrafast ionization of neutral clusters at the leading edge of the FEL pulse. We have shown that the method is sensitive to the target composition. The accurate modeling of a target ionization in the focus volume [46] is complicated by significant shot-to-shot fluctuations of almost all FEL pulse parameters such as pulse energy and pulse duration as well as spatial intensity distribution [1,2]. In our study we show the applicability of the THz-field streaking technique to temporally resolve the electron emission from extended targets under intense FEL radiation on time scales that are significantly shorter than the FEL pulse duration. This is a regime in which NIR (or attosecond) streaking cannot be applied, because the required NIR intensities would lead to additional ionization of the clusters. This can be avoided by using a THz pulse with a much lower intensity, but with a similar ponderomotive energy. We expect that our results will stimulate further development of the theory of the light-field streaking which considers the multielectron dynamics in highly excited systems. With continuous development of THz sources that are able to provide intense single or multicycle THz pulses [47], these types of experiments will significantly advance the ultrafast x-ray physics at existing and emerging state-of-the-art user facilities [1–3] as well as in the laboratory [48].

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