Direct autocorrelation of soft-x-ray free-electron-laser pulses by time-resolved two-photon double ionization of He

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The pulse duration of soft x-ray free-electron laser (FEL) radiation is directly measured by time-resolved observation of doubly charged helium ions at 51.8 eV. A wave front splitting autocorrelator produces two correlated FEL pulses with a resolution of better than a femtosecond. In the interesting intensity range from 10^{13} to 10^{16} W/cm^2 direct and sequential double ionization contribute to the ion yield which has significant influence on the correlation width, being a general feature at high photon energies. Here, a duration of $\tau_L = (29 \pm 5)$ fs is derived for the soft x-ray pulses at FLASH.

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Nonlinear optical processes have tremendously advanced the knowledge about the electronic structure of matter, and enabled ultrafast time-resolved investigations of dynamic processes by various correlation techniques. Meanwhile such processes have abundantly been studied employing coherent laser sources in the visible and uv spectral regime. At soft x-ray photon energies, however, the lack of intense laser sources prohibited so far detailed investigations of multiphoton excitations. Recently, two-photon above threshold ionization [1] and two-photon double ionization [2] of He atoms have been observed with coherent soft x-ray radiation obtained from high harmonic generation.

Advances in free-electron lasers operating in the self-amplified spontaneous emission (SASE) mode open a new spectral window for nonlinear studies in the soft x-ray range [3]. The interaction of this intense soft x-ray radiation with the electron clouds of atoms, molecules, clusters, and condensed matter systems is expected to cause new and hitherto not yet uncovered nonlinear electronic phenomena through the many particle nature of these processes at high photon energies. In recent, not time-resolved experiments indications of the existence of such interactions are found, such as the observation of Xe^{21+} produced with 93 eV radiation which requires the absorption of more than 50 soft x-ray photons [4] or the multiple step ionization of Ar clusters with 38 eV radiation which differs significantly from processes at lower photon energies [5]. In these experiments intensities up to a few times 10^{15} W/cm^2 in the soft x-ray regime have been applied. At somewhat lower intensities electron correlation effects have been observed in the few-photon ionization of Ne and Ar where nonsequential two-photon excitation and at higher intensities nonsequential and sequential three-photon processes contribute [6]. The theoretical understanding of such electronic multiphoton processes at high photon energies poses also interesting questions, especially when the photon energy is larger than required for driving the system above the first few ionization steps. The contributions of different processes will depend upon the specific atomic or molecular system and on the characteristics of the soft x-ray radiation, such as pulse duration, intensity, and coherence. For the understanding of such nonlinear processes as well as for the determination of absolute photoionization cross sections [7] precise knowledge of the duration and time structure of the high-intensity soft x-ray pulses applied is of utmost importance. So far these important time characteristics of soft x-ray free-electron laser pulses have only been estimated from electron bunch simulations [8]. In this Brief Report we report on the first determination of the average pulse duration of the SASE free-electron laser at Hamburg (FLASH) by direct autocorrelation in the soft x-ray regime via time-resolved generation of doubly charged He ions at a photon energy of $h\nu = 51.8$ eV.

Early calculations for the two-photon double ionization of He, the simplest system in which electron correlation can be

expected, reveal a variation of the two-photon cross section for direct double ionization within nearly one order of magnitude [9–11]. For photon energies below the ionization threshold of He^+ (54.4 eV) Lambropoulos and co-worker found that above a certain intensity the sequential double ionization yield dominates the direct double ionization yield [12,13]. The intensity where this occurs depends on the photon energy used and the duration of the pulses applied, and it shifts to higher values for higher photon energies and shorter pulses. This behavior is caused by the fact that for photon energies between about 38 and 54 eV the sequential process is of third order while the direct one is a two-photon process. The direct process, therefore, dominates the ion yield at intensities below about 8×10^{14} W/cm^2 at 51.8 eV.

Both pathways, however, involve a nonresonant two-photon process contributing to the delay-dependent generation of He^{2+} and are therefore well suited to measure a nonlinear autocorrelation trace to determine the duration of ultrashort pulses in this spectral range. The different intensity dependencies of these nonlinear pathways might create artifacts in the correlation curves, a problem of increasing importance for still higher photon energies where nonlinear autocorrelation in multiply charged ions is envisioned and even multiple pathways may contribute. Therefore, we specifically address their influence on the correlation width.

In this experiment, time-correlated soft x-ray laser pulses are produced by a wave front splitting device [14]. The two partial beams from the soft x-ray beam splitter are recombined by a normal incidence spherical Mo/Si multilayer mirror with a focal length of 20 cm and a surface figure of $\lambda/20$. This mirror shows a maximum reflectivity of about $R=30\%$ at 24.0 nm and a spectral width of 1.5 nm [full width at half maximum (FWHM)] ($\Delta E\sim3.2$ eV). The combined beams are estimated to be focused to a focal diameter about 5 μm. FLASH operated in the pulse energy range of 7–11 μJ. The pulse-to-pulse intensity fluctuations were about 40% rms. A total transmission in both arms of the autocorrelator of 48% is measured at 23.9 nm (51.8 eV) using two gas monitor detectors [15] in front and behind the beam splitter. Taking the transmission of the beamline (70%), the beam splitter and the mirror reflectivity into account soft x-ray peak intensities of about 1.8×10^{14} W/cm^2 are applied. He^{2+} ions produced by the FEL pulses in the ionization region of a Wiley-McLaren-type time-of-flight mass spectrometer are extracted by a homogeneous electric field through a small slit with a width of about 350 μm and a height of 1 mm, see Ref. [4]. This ensures that only ions from the focal region are detected while background ions from the unfocused beam are rejected. The extracted ions are detected by an open multiplier. The signal of the multiplier is registered by a fast digital oscilloscope and stored in a computer.

Prior to and immediately after the time-resolved two-photon ionization experiment the zero delay of the autocorrelator setting is determined by measuring the linear two-beam interference pattern generated by overlapping both partial beams under a small angle of $\alpha=0.18$ mrad directly on a soft x-ray sensitive charge-coupled device (CCD) camera. This yields a temporal coherence of the FEL pulses at FLASH of about $\tau_{coh}=6$ fs (FWHM) [16], exactly at this wavelength and measured during the same beam time. In this context the maximal visibility indicates the time zero in the delay of the two beams. It is worth noting that this maximal coherence occurs at the same delay settings of the autocorrelator as the maximal nonlinear ionization signal of He^{2+}.

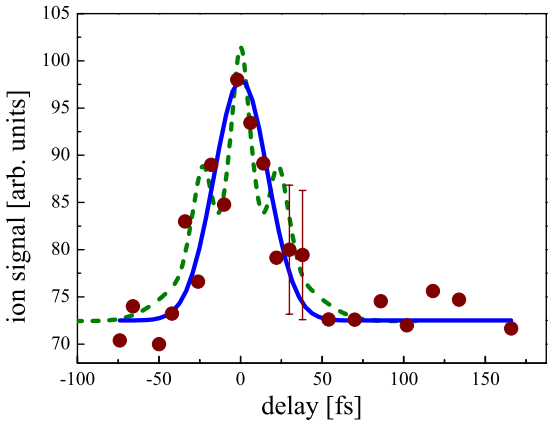

FIG. 1. (Color online) Time-resolved double ionization of He (dots). The solid line denotes a Gaussian function with 39 fs FWHM yielding a pulse duration of $\tau_L=(29\pm5)$ fs, the dashed line represents a three-pulse structure with temporal separations of the maxima of 12 and 40 fs and an added chirp of 50 fs^2 as inferred from Ref. [16].

Figure 1 shows the He^{2+} yield observed (dots) as a function of delay time between the two beams measured at a partial pressure of about 5×10^{-5} mbar. The average of several independent delay scans is shown over a time interval of 250 fs. Each data point of the individual scans is averaged over 500 FLASH pulses which operated at 5 Hz repetition rate. It is evident that a significant increase of the He^{2+} signal is observed when the pulses from the two partial beams overlap in time. The error bars indicate the statistical signal variation of the individual scans. The solid line shows a Gaussian fit to the experimental data. This yields an autocorrelation width of about 39 fs. Under the assumption of a Gaussian shape a pulse duration of $\tau_L=(28\pm5)$ fs would then be derived.

The dashed line shows the nonlinear intensity autocorrelation obtained using the three-pulse structure inferred from the previous linear autocorrelation measurement with two main pulses separated by 12 fs and a weak trailing pulse 40 fs behind the first one [16]. In addition, a linear chirp of 50 fs^2 is assumed which easily stretches the Fourier limited subpulses to the pulse duration observed. This pulse structure, previously only inferred from the nonmonotonous coherence function and observed during the same run of the FEL, would therefore also be compatible with the nonlinear autocorrelation. Due to the scatter of the data a clear distinction between both fits is currently not possible.

It may be noticed that the peak-to-background intensity ratio deviates from the value of 3:1 usually expected for an intensity autocorrelation. Such a ratio is obtained only for truly collinearly propagating beams. In the present case, however, the two partial beams emerge side-by-side from the autocorrelator with an initial separation of about one mm, and are therefore focused in a crossed-beam geometry. Fol-

lowing a ray tracing analysis and including imaging errors, as witnessed by the much larger than diffraction limited focal spot size, this already yields a ratio of only about 2:1 for otherwise perfect spatial overlap. Additionally, pulse-to-pulse fluctuations of the focal plane position due to a variation of the source point of the SASE radiation in the undulator as well as an imperfect vertical beam overlap cause a further reduction of the delay-dependent signal. These effects lead to a significantly smaller peak on top of the delay independent He^{2+} ion signal from the two single beams. A refined analysis of the spatial distribution of the electric field in the ionization volume, as performed by Tzallas and co-workers [17] for the characterization of attosecond pulse trains, is not expected to yield a better estimate of this ratio, because of the strong pulse-to-pulse fluctuations not only in intensity but also in the beam profile of SASE free-electron laser pulses like FLASH. Electro-optic sampling and transverse deflecting radiofrequency measurements of the electron bunches at FLASH operating in the femtosecond mode recently reveal an electron bunch duration in the range between 30 to 55 fs [18], thus well within the present scan range.

The observed pulse duration of about 29 fs for averaged SASE FEL pulses is significantly longer than the temporal coherence of about 6 fs [16]. This can be rationalized by concluding that the FEL pulses consist of more than a single coherent emission event, as is expected from a FEL operating in the SASE mode [8]. Single shot spectra as well as linear autocorrelation measurements taken during the same run, see Ref. [16], strongly support this conclusion. Due to the energy chirp of the lasing part of the electron bunch [3], it is expected that these subpulses are not Fourier transform limited, but show a spectral phase modulation. In principle, such a spectral phase modulation may also be acquired from the reflections of the soft x-ray radiation on the beam line mirrors and those within the autocorrelator. However, all these reflecting mirror surfaces consist of diamondlike amorphous carbon layers with an attenuation length of typically 4.1 nm at 51.8 eV [19]. From the spectral dependence of this penetration depth the totally acquired pulse broadening after eight reflections amounts to only a few attoseconds. Also the Mo/Si focusing multilayer mirror contributes at most an estimated 10 to 20 as to the pulse duration. A pulse lengthening due to the mirror reflections can therefore be neglected.

The ionization dynamics and their influence on the nonlinear autocorrelation has been modeled using the electric field strength of the pulses with a set of rate equations similar to that used earlier [9,12,13]. The direct nonsequential double ionization cross section $\sigma_a(\mathrm{He}\rightarrow\mathrm{He}^{2+})$ of He has recently been calculated by Feist *et al.* [20], Guan *et al.* [21], and Ivanov and Kheifets [22] in the photon energy range from about 40 to 54 eV. At low intensities their results are in agreement with experimental values at 41.8 eV [23] and 42.8 eV [7] and other recent theoretical calculations, see references in [13,21]. At $h\nu=51.8$ eV a value of $\sigma_a=2.0\times10^{-52}$ cm^4 s is taken for the direct path in the model calculation discussed below. As an alternative route the sequential double ionization is considered. Here first a single photon ionization of He to $\mathrm{He}^+(1s)$ with a cross section of $\sigma_b(\mathrm{He}\rightarrow\mathrm{He}^+)=1.90\times10^{-18}$ cm^2 occurs [13,24], followed by a nonresonant two-photon ionization of $\mathrm{He}^+(1s)$. This last cross section can exactly be calculated since it constitutes a single electron hydrogen problem showing a two-photon ionization cross section of about $\sigma_c(\mathrm{He}^+\rightarrow\mathrm{He}^{2+})\sim1.5\times10^{-53}$ cm^4 s [13], averaged over the spectral width of the free-electron laser pulse.

From the rate equation model it turns out that up to a peak intensity of a few times 10^{14} $\mathrm{W/cm}^2$ and a pulse duration of about 30 fs the direct double ionization dominates the pro-

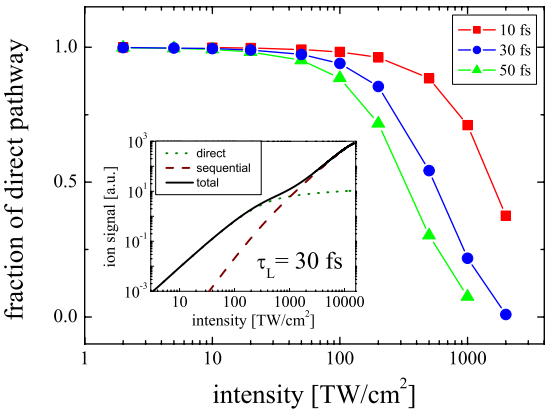

FIG. 2. (Color online) Contributions of the direct and sequential pathways to the total He^{2+} double ionization signal as a function of peak intensity for Gaussian pulses of different durations. Note that here the intensity scale shows the intensity of just one partial beam. The inset shows the intensity dependence of both pathways for 30 fs long pulses.

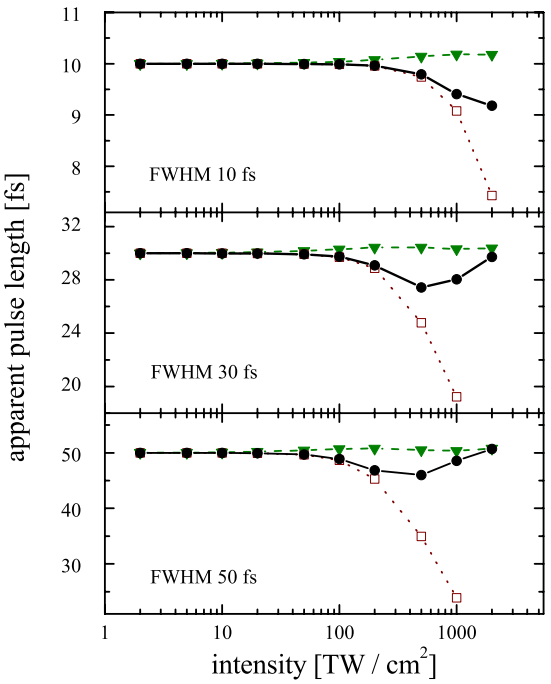

FIG. 3. (Color online) Influence of the pulse intensity on the autocorrelation trace at 51.8 eV for pulse durations of 10, 30, and 50 fs. The open squares and filled triangles represent apparent pulse widths when the direct and sequential double ionization path are analyzed separately. The filled dots show the pulse widths when the total He^{2+} signal is measured.

duction of He^{2+} ions, see Fig. 2. The He^{2+} signal observed in this study at about 1.8×10^{14} W/cm^2 thus results dominantly (94%) from a single nonresonant two-photon process. It is therefore safe to derive the corresponding pulse width from the correlation curve measured. At higher intensities or longer pulses the neutral helium is strongly ionized already by the rising wing of the FEL pulse. Consequently, the population of neutral He in the beam volume is dramatically reduced. Therefore, at intensities larger than 10^{15} W/cm^2 and a pulse duration of 30 fs or longer the generation of He^{2+} proceeds dominantly via two-photon ionization of He^+, in agreement with earlier theoretical yield calculations at lower photon energies [12,13].

In general, the depletion of the neutral ground state and the presence of two channels to generate He^{2+} may influence the autocorrelation curve up to the point that a meaningful extraction of a pulse duration may no longer be possible. Figure 3 shows the apparent pulse durations derived from an autocorrelation trace of He double ionization for nominally 10, 30, and 50 fs long Gaussian pulses as a function of peak intensity. The open squares and filled triangles show the durations derived from separately analyzing the direct and sequential double ionization, respectively, while the filled dots show pulse durations when observing only the total He^{2+} signal, as in this investigation. It is evident that for increasing intensities the direct double ionization path leads to seemingly shorter pulses, especially for longer pulse durations. This is caused by the depletion of the neutral He ground state. Above a few times 10^{15} W/cm^2 this channel does practically not contribute anymore to the He^{2+} yield. On the other hand, the sequential double ionization pathway shows a small but negligible broadening, because first He^+ ions have to be generated which takes some fraction of a femtosecond. This signal is nearly not affected even at high intensities above 10^{15} W/cm^2. The total He^{2+} signal only slightly underestimates the true pulse duration by up to 10% in the intensity region from 10^{14} to 10^{15} W/cm^2. At the intensities used in this study the true pulse duration is thus underestimated by less than 1 fs. In conclusion, from a time-resolved measurement of He double ionization we deduced a pulse duration of about $\tau_L=(29\pm5)$ fs for the SASE free-electron laser at FLASH operating in the femtosecond mode at a photon energy of 51.8 eV. An analysis of the contributions of different ionization pathways is necessary in order to arrive at meaningful conclusions about the true pulse duration. This sensitivity of the correlation width on the pathway will be even more important when other gases and higher ionization states are used to characterize pulse durations at higher photon energies. Together with a concurrent determination of the coherence time of 6 fs [16] a substructure of the femtosecond FEL pulses is very likely.

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