Femtosecond silicon $K\alpha$ pulses from laser-produced plasmas

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Ultrashort bursts of silicon $K\alpha$ x-ray radiation from femtosecond-laser-produced plasmas have been generated. A cross-correlation measurement employing a laser-triggered ultrafast structural change of a CdTe crystal layer (320 nm) shows a $K\alpha$ pulse duration between 200 fs and 640 fs. This result is corroborated by particle in cell simulations combined with a Monte-Carlo electron stopping code and calculations on the structural changes of the crystal lattice.

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Since their discovery in 1895, x rays have had an enormous impact on the development of science. They are used to obtain structural information on any type of matter on an atomic scale. Combined with pulsed lasers, synchrotrons, and x-ray streak cameras, even time resolved x-ray studies with a resolution of some picoseconds have become possible. Laser-induced crystal strain, for example, has been successfully investigated with picosecond time resolution using x-ray pulses produced by picosecond lasers [1] and synchrotron radiation combined with streak cameras [2]. Ultrashort laser pulses have opened a new dimension for these studies since they offer the possibility to create intense bursts of incoherent energetic x rays [3]. Moreover, simulations predict that the pulse duration is of the order of a few hundred femtoseconds [4]. This has created the possibility of time resolved x-ray diffraction on a picosecond or even a sub-picosecond time scale [5–10]. In these experiments, $K\alpha$ radiation was used to monitor ultrafast structural changes in matter. An open question in all of these pump-probe experiments was, however, the exact temporal duration of the probing x-rays. This ultimately determines the temporal resolution of the experiment. The determination of the temporal duration of sub-picosecond x-ray pulses is very challenging. X-ray streak cameras have in practice a temporal resolution of 0.5 ps for single-shot mode and about 3 ps for multishot mode [11,2]. Other experimental methods developed for IR, visible, and UV pulses are related to autocorrelation techniques and may not easily be extended into the x-ray regime. The basic reason for this difficulty lies in the fact that nonlinear optical effects that provide the essential experimental tools at larger wavelengths have so far not been observed in the x-ray regime. Therefore, other methods that cross correlate a visible laser pulse of known temporal duration with x-ray pulses have to be used. Several successful experiments that employ such methods in the VUV and XUV spectral region have been reported [12–14]. Measurements of the temporal duration of pulses consisting of multi-kilovolt photons—the regime of interest for temporally resolved structure analysis—have, however, not yet been performed.

In this paper we report on the measurement of the time duration of the Si $K\alpha$ line emission from a femtosecond-laser-produced plasma. The measurement is performed by cross correlating the Si $K\alpha$ pulse with the ultrafast response of a laser pumped MBE-grown thin CdTe crystal layer. In contrast to InSb bulk measurements [10], the x-ray penetration depth in this measurements is definitely limited by the thickness of the crystalline layer. The pump laser induces an ultrafast change of the CdTe crystal structure and causes a rapid decrease of the x-ray reflectivity. We have performed combined particle in cell (PIC) and Monte Carlo electron propagation simulations in order to estimate the Si $K\alpha$ pulse width. In addition, the response of the CdTe crystal layer was simulated by assuming a random displacement of the ions in the crystal lattice [15]. The numerical results are in good agreement with the experimental data.

In order to measure the temporal evolution of the $K\alpha$ burst a cross-correlation technique was employed. The basic idea of the measurement is to reflect the x-ray pulse from a CdTe crystal layer to a charge-coupled-device (CCD) camera. A second laser pulse synchronized to the plasma-creating pulse is focused on the surface of the CdTe crystal layer and destructs the uniform crystal structure within a depth comparable to the absorption depth of the laser pulse. The reflectivity of the undisturbed crystal is normalized to $R=1$. If the reflection takes place after the crystal surface has been modified, the reflectivity decreases to a value that is mainly determined by the ratio of the thickness of the modified layer and the penetration depth of the x-ray radiation. The absorption depth of the Si $K\alpha$ and the laser radiation in CdTe is 498 nm and (175 ± 15) nm, respectively. Assuming a modified surface layer with a thickness equal to the penetration depth of the laser radiation, the reflectivity of the 320 nm thick CdTe crystal layer should decrease to $R=0.53$. Varying the delay between the two pulses and analyzing the transition region allows us to deduce the temporal duration of the x-ray pulse, presumed the time constant of the crystal response is known.

The experiment setup is shown in Fig. 1. The experiments were performed with a chirped pulse amplification (CPA) Ti:sapphire laser system delivering 80 fs pulses with a maximum energy of 200 mJ at a repetition rate of 10 Hz. The contrast ratio of the laser has been measured using a fast photodiode (ns regime) and a third order background-free autocorrelator (fs to ps regime). Prepulses were found at 7 ns and 4 ps with an intensity contrast ratio of $>10^6$ and 10^4,

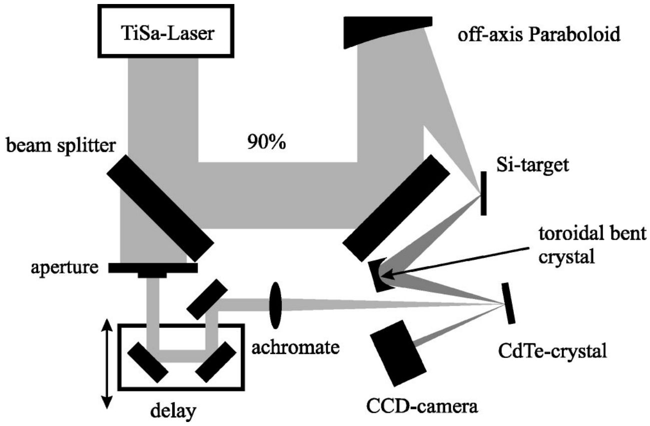

FIG. 1. The Ti:sapphire laser system delivers pulses of about 80 fs with a pulse energy of 200 mJ. 90% of the energy is focused onto a silicon target by an off-axis paraboloid, producing a short burst of Si $K\alpha$ line emission. A fraction of the 10% transmitted laser energy is used to induce changes in the lattice properties of the CdTe crystal layer. The changes are probed by the silicon $K\alpha$ pulse and detected by an x-ray CCD camera.

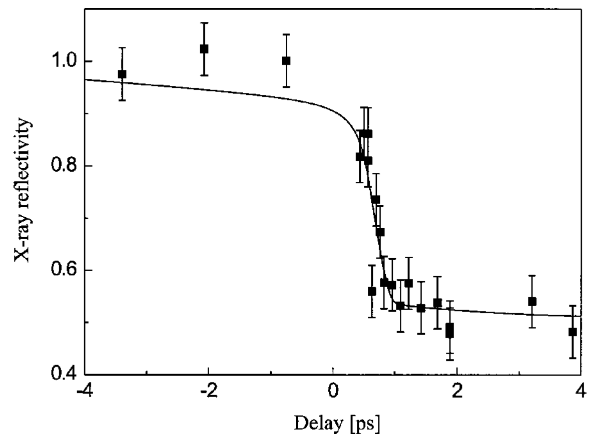

FIG. 2. The reflected x-ray intensity as a function of the delay time between the optical pump (3.2 J/cm^2) and the x-ray probe pulse (negative delay times: x-ray probe before optical pump). The reflectivity decreases on a time scale of several hundred femtoseconds to (0.51 ± 0.02). The solid line corresponds to the best fit using the calculated x-ray pulse and yields a decay of the x-ray reflectivity with a time constant of 540 fs (linear decay, 90% to 10%).

respectively. In the target chamber (pressure 10^{-4} mbar) the incoming laser pulses were split by a beam splitter with a ratio of 9:1. The main part of the pulse (90%) was focused on a silicon target with a spot size of 10×100 μm^2 corresponding to a maximum intensity of 3×10^{17} W/cm^2 (p polarization). Fast electrons are produced during the laser plasma interaction and their average energy is of the order of a few tens of keV [16–19]. Since the interaction time of the laser electric field with the electrons is limited to the laser pulse duration, it is expected that the temporal width of the electron burst is comparable to the laser pulse width. The electrons subsequently penetrate into the bulk material behind the plasma and produce characteristic line emission [17,19]. An x-ray spectrum was recorded between 6.6 Å and 7.2 Å with an absolutely calibrated Pentaerytrit von-Hamos spectrometer [20]. Integrating the $K\alpha$ line yields 6×10^9 photons per shot within a solid angle of 4π. An additional angle-dependent measurement showed that the line emission is isotropic within a half sphere. The $K\alpha$ radiation was refocused onto a 320 nm thick single crystal CdTe(111) layer (on a GaAs substrate) by using the 10.0 reflection of a toroidally bent quartz. The diameter of the x-ray focus was 38 μm. Because the source was located on the Rowland circle the spectral width of the reflected x-ray radiation is determined by the rocking curve of the bent crystal and the size of the x-ray source to $\Delta\lambda/\lambda = 10^{-4}$. A CCD camera was used to record the Bragg reflection (72.34°) from the CdTe crystal layer. The line width is determined by the intrinsic reflection curve of the CdTe crystal layer (3.3 mrad) and again by the size of the $K\alpha$ source (0.6 mrad).

The remaining 10% of the laser pulse transmitted through the beamsplitter are used to change the CdTe crystal structure in order to realize an ultrafast x-ray gate. A variable delay line alters the time delay between the plasma generating and the optical pump pulse. The spatial overlap between the optical pump and the x-ray probe pulse is critical for the experiment and had to be carefully aligned. The temporal overlap between both the pulses was determined by spectral interferometry. To perform this measurement, the plasma generating pulse was attenuated so that no plasma was produced and the laser pulse was reflected towards the bent x-ray crystal. An overlap with the optical pump pulse at the position of the CdTe crystal layer and imaged onto the entrance slit of a spectrometer allows to be adjusted the zero delay between the two pulses.

Figure 2 shows the reflected x-ray intensity from the CdTe crystal layer as a function of the delay between the optical pump and the x-ray probe pulse. Clearly, the reflectivity decreases to $R = (0.51 \pm 0.02)$ in good agreement with the predictions. Even for ''infinitely'' long delay times (about 10 min) the reflectivity remains at $R = (0.51 \pm 0.02)$ indicating that a permanent modification of the CdTe crystal structure took place. Most important, the decay of the reflectivity from 0.95 to 0.55 corresponding to a change from 90% to 10% (linear decay) of the signal occurs within (640 ± 50) fs. The geometry of the experimental setup, i.e., the size of the x-ray emitting region, the tilt between the optical pump and the x-ray probe beam, and the finite penetration of the Si $K\alpha$ radiation in the focusing crystal, leads to a total system response time of 30 fs. Therefore, systematic errors are negligible as compared to the observed time constants. The result may be interpreted in two extreme ways. First, the transition from the reflecting to the nonreflecting state of the CdTe crystal layer happens within an infinitely short time, then a full width at half maximum (FWHM) of the x-ray pulse of (640 ± 50) fs is inferred. Conversely, if the x-ray pulse width is assumed deltalike, the crystal transition occurs within (640 ± 50) fs. To visualize this analysis, a x-ray pulse was cross correlated with a linearly decaying CdTe crystal response. Figure 3 shows the quadratic deviation between the computed curve and the measured data as a function of the FWHM of the x-ray pulse and the decay time of the linear response from 90% to 10%. The dark region indicates the most likely response time for a given x-ray FWHM. Earlier

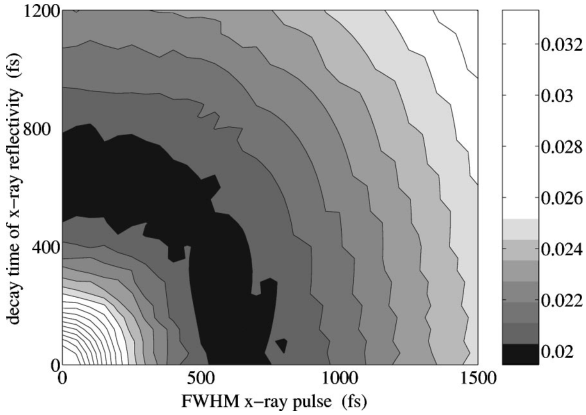

FIG. 3. Quadratic deviation of the correlation of different Si $K\alpha$ pulse durations (FWHM) with different decay times (linear decay, 90% to 10%) of the CdTe x-ray reflectivity from the experimental data (Fig. 2).

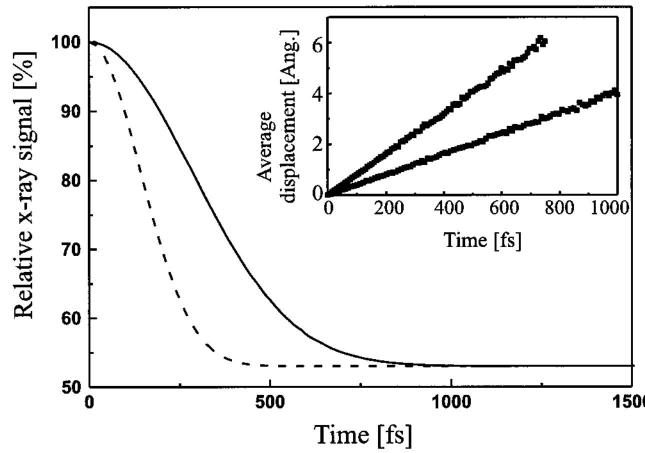

FIG. 5. Assuming a randomly oriented motion of the Cd and Te atoms with an average displacement velocity of 4 Å /ps (solid line) to 8 Å /ps (dashed line) following laser irradiation allows us to calculate the reflectivity ($\propto\langle F_{hkl}\rangle^2$, where $\langle\cdots\rangle$ denotes the ensemble average) of the CdTe crystal layer as a function of time. The inset shows the average displacement as a function of time for 4 and 8 Å /ps, respectively.

measurements are in agreement with this analysis [9]. In this paper the number of data points is significantly higher and represents a much more precise determination of the decay of the reflectivity. Nevertheless, both measurements depend on the transient behavior of the different crystalline materials and neither show any discrepancy with the result of Fig. 3.

For further evaluation of the $K\alpha$ pulse duration we have performed simulations of the Si $K\alpha$ generation and the transient change of the CdTe crystal structure.

A PIC code was used to calculate the energy distribution of the electrons for the given laser parameters. Then, a Monte Carlo code was employed to extract the $K\alpha$ yield and the temporal evolution of the $K\alpha$ emission. Figure 4 shows the calculated FWHM of the Si $K\alpha$ radiation as a function of the laser intensity. In contrast to previously investigated higher Z materials [4] the FWHM is almost independent of the laser intensity (175 fs to 195 fs). In principle, the pulse width should increase with laser intensity because the mean electron energy increases with intensity and consequently the penetration depth of the electrons into the bulk material increases. In the case of silicon the absorption length of Si $K\alpha$ in silicon bulk material is of the order of 12 μm, which confines the $K\alpha$ emitting volume to a depth comparable to the $K\alpha$ penetration depth. Radiation produced by electrons that penetrate deeper into the bulk cannot escape due to absorption. This in turn sets the upper limit for the $K\alpha$ pulse prolongation due to the increasing penetration depth of the electrons to about 140 fs. In conclusion, the calculations predict a $K\alpha$ pulse duration between 175 fs and 195 fs for a laser intensity in the range of $[0.7–3]\times10^{17}$ W/cm^2.

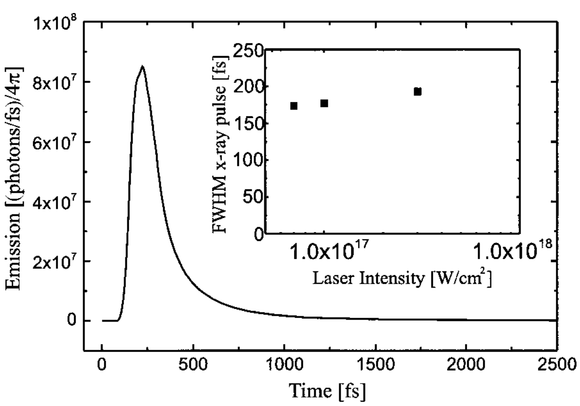

FIG. 4. Calculated temporal intensity profile of the Si $K\alpha$ pulse for an intensity of 10^{17} W/cm^2. The inset shows the calculated FWHM as a function of the incident laser intensity.

The physical processes triggered by fs-laser irradiation of the CdTe crystal may be explained in the following way. The pulse energy is coupled into the electron-hole plasma and subsequently transferred to the ions by electron-ion collisions until thermal equilibrium is reached. If a fluence below a critical threshold is used, the heating of the electrons does not lead to structural changes before electrons and ions have equal temperature. All subsequent processes are thermal and happen on time scales that are determined by the characteristic velocities of the thermal processes. Once the fluence exceeds a certain threshold, the number of generated electron-hole pairs becomes so high that the binding forces between the ions are substantially disturbed. The ions start to move away from their equilibrium positions and the structural order is deranged on a time scale of a few hundred femtoseconds [7,21–23]. Allen *et al.* [15] have calculated an average displacement velocity of about 8 Å /ps in the case of InSb and 4 Å /ps in the case of GaAs. We have estimated the decay of the x-ray reflectivity assuming that the atoms move in random directions once the electron-hole plasma destabilizes the crystal structure. The velocities for the Cd and the Te atoms were varied between 4 and 8 Å /ps. Figure 5 shows the calculated x-ray reflectivity as a function of time, assuming that the thickness the modified region is equal to penetration depth. Clearly, the reflectivity decreases from 90% to 10% within 240 fs to 480 fs depending on the average displacement velocity. If we use the calculated x-ray pulse, the best fit to the experimental data leads to a decrease of the x-ray reflectivity with a time constant of 540 fs (linear decay, 90% to 10%). This in turn would indicate that the average displacement velocity is comparable to the value calculated for GaAs.

In conclusion, with an optical pump and a x-ray probe arrangement the temporal duration of the Si $K\alpha$ emission from a femtosecond-laser-produced plasma has been measured. From the experimental data we obtain an upper limit of 640 fs for the Si $K\alpha$ pulse duration assuming an infinitely fast structural change of the CdTe crystal layer and a lower limit of 200 fs from the simulations. The most probable value for the $K\alpha$ pulse duration is therefore between 200 fs and 640 fs. In contrast to previously published experiments [5,7,9,10], this work considers not only the crystal response but also the x-ray emission duration. We have deconvolved the experimental curve (Fig. 2) with the calculated $K\alpha$ pulse shape (Fig. 4) and obtain a temporal change in the CdTe $K\alpha$ reflectivity that corresponds to an average displacement velocity (Fig. 5) close to 4 Å /ps, which is similar to GaAs. Experiments and simulations show, however, that the $K\alpha$ pulse duration and the time scale for a structural change in the thin CdTe crystal layer are of roughly the same magnitude. Since the atomic displacement velocities in other ultrafast x-ray diffraction experiments reported recently [5–10] are comparable to those of CdTe, similar limitations on the temporal resolution would also apply to those experiments.

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