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Temporal probing of an ultrafast plasma shutter driven by a KrF femtosecond laser system

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Abstract. A femtosecond high-power KrF-laser system was used to create a rapidly ionizing plasma on a BK7 glass slide and the plasma formation was probed by a second laser pulse at 497 nm. During the formation of the plasma the electron density reaches the critical density for 497 nm. Therefore, the reflectivity of the surface rises from a value given by the Fresnel formulas to a value close to 100% and the transmission decreases on the same time scale. It is shown that this method permits a precise timing of two femtosecond laser pulses at different wavelengths.

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Pump-probe experiments in the ultrashort pulse laser regime often require to overlap two pulses of different wavelengths in time. Since the temporal resolution of all electronic devices and even the fastest streak cameras is too poor in order to adjust the temporal overlap, cross-correlation techniques have to be used. Cross-correlation methods, however, require a nonlinear medium, which in some spectral ranges is hard to find. In the following report a fast optical gate based on the rapid ionization of a transparent glass surface is presented. It is shown that the plasma formation happens on a time scale similar to the pulse width of the plasma creating pulse. The energies required are on the order of a few tens of μJ. This optical gate is, in principle, independent of the wavelength and, therefore, can be used to overlap two pulses at almost arbitrary wavelengths using a relatively simple experimental setup.

In the following the experimental setup is described. A model is presented which allows for a qualitative understanding of the experimental results. It is shown that under certain constraints the method is suitable for measuring the temporal duration of fs laser pulses. Since the decrease of the electron density is governed by hydrodynamic expansion of the plasma, the gate has a long lifetime compared to the temporal width of the laser pulses. Therefore, the gate is a step-like function and a very useful tool to overlap two ultrashort laser pulses with different wavelengths in time.

1 Experimental setup

The experimental setup is shown in Fig. 1. The XeCl-excimer-laser-pumped dye laser system delivered pulses with a temporal duration of about 400 fs at a wavelength of 497 nm [1]. Part of the VIS pulses was frequency doubled by a 300-μm-thick BBO crystal and injected into a KrF-excimer amplifier tube. After amplification the UV pulses had an energy of about 12 mJ. The pulse width was about 700 fs (FWHM) due to linear and nonlinear effects in the amplifier. Using a prism compressor consisting of two CaF_2 prisms the UV pulses could be compressed to a minimum pulse width of about 200 fs. The total transmission of the prism compressor was 45%. The remaining part of the pulses at 497 nm was coupled into a polarization-preserving monomode fiber with a core diameter of 1.5 μm and a length of 33 cm. The spectral width of the pulses increased from about 1 nm to typically 10 nm due to self-phase modulation (SPM) in the fiber. After the fiber the pulses were amplified by two excimer-laser-pumped dye cells (Coumarin 307) to a maximum energy of 40 μJ. The subsequent compression was realized by a prism compressor consisting of two SF10 prisms. For ideal conditions the pulses emerging from the fiber should have a nearly linear phase modulation [2]. Therefore, a spectral width of 10 nm should support a pulse width of about 40 fs. However, due to residual phase modulations, which could not be compensated with the prism compressor, the minimum pulse width obtained was about 90 fs. The UV pulses at 248 nm were characterized using a frequency-resolved optical gating (FROG) setup. The nonlinear medium was a 1.5-mm-thick plate of fused silica [3]. In the inset of Fig. 2 an auto-correlation trace derived from the measured FROG trace is shown. From this a pulse width of the UV pulses at 248 nm of (215 ± 10) fs is obtained. The laser pulses at 497 nm were measured by a multi-shot second-harmonic generation FROG [4]. Figure 2 shows the FWHM of the laser pulses at 497 nm as a function of the separation of the two SF10 prisms. The minimum pulse width of 90 fs was obtained at a prism separation of about 64 cm. The UV pulses were spatially combined with the pulses at 497 nm by a di-

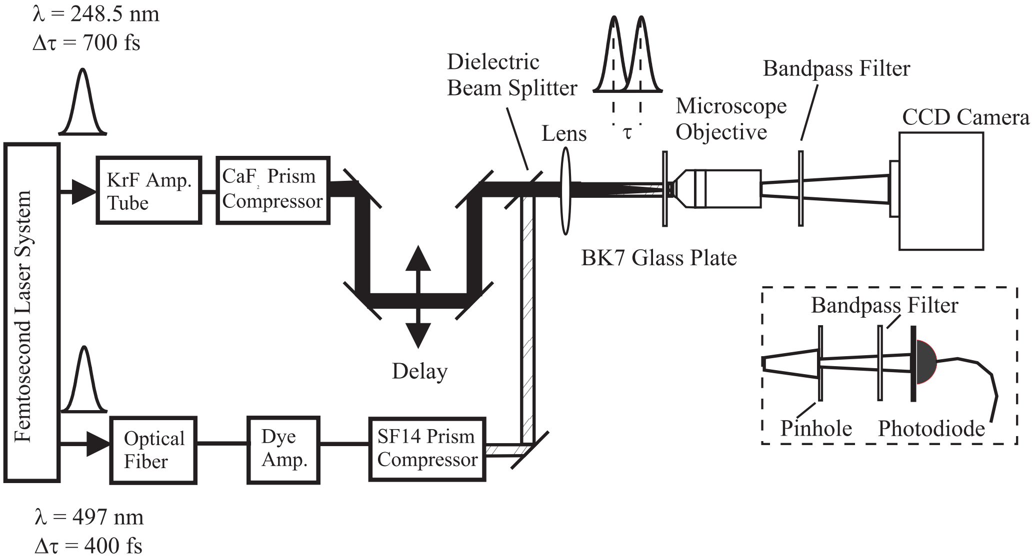

Fig. 1. The fs dye laser system delivered pulses of about 400 fs FWHM at a wavelength of 497 nm. Part of the pulse was frequency doubled and subsequently amplified in a KrF amplifier tube. The UV pulses were compressed to about 160 fs by a prism compressor (CaF_2). The remaining part of the pulses at 497 nm was focused into a monomode fiber and the spectral bandwidth increased from about 1 nm to typically 12 nm. The pulses emerging from the fiber were amplified and compressed by a prism compressor (SF10) to about 90 fs

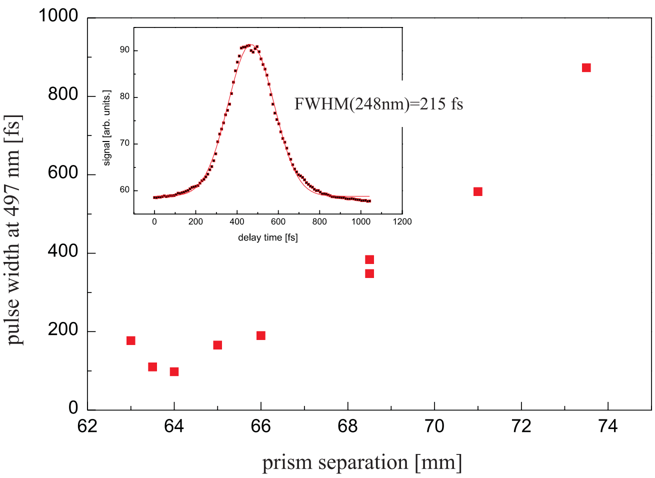

Fig. 2. The temporal width of the pulses at 497 nm (FWHM) is shown as a function of the separation of the two SF10 prisms. The minimum pulse width of about 90 fs is obtained at a prism separation of 64 cm. The *inset* shows a third-order auto-correlation trace of the compressed pulses at 248 nm. The auto-correlation trace was obtained from a single-shot FROG setup and a pulse width of (215 ± 10) fs is inferred

electric beam splitter. Both pulses were focused collinearly onto the sample (1-mm-thick BK7 glass plate) using a plano-convex fused silica lens with a focal length of 50 mm. The focus on the front surface of the sample was imaged onto a CCD camera in transmission geometry using a ×10 microscope objective (see Fig. 1). The CCD camera was equipped with a dielectric bandpass filter with a center wavelength of 497 nm in order to reject fluorescence light from the laser-produced plasma. The UV light was very efficiently absorbed by the BK7 glass plate.

The energy of the UV pulses impinging onto the glass plate was (150 ± 50) μJ and the focal spot was (50 ± 5) μm in diameter leading to a peak intensity of about 3.6×10^{13} W/cm^2. The energy of the green pulse was on the order of 1 μJ and the beam diameter on the glass plate about 250 μm. Therefore, the intensity of the pulses at 497 nm was on the order of 10 GW/cm^2. This intensity is too low to create a plasma on the BK7 target. The relative delay between the two pulses was adjusted by a translation stage with an accuracy of 2 μm, i.e. 13 fs. Negative delay times correspond to situations were the pulses at 497 nm precede the pulses at 248 nm. The samples were mounted on a *x*-*y* translation stage perpendicular to the laser beams and were moved between two subsequent laser pulses in order to ensure that the pulses always interacted with an intact surface. Figure 3a shows a typical image of the pulse at 497 nm for negative delay times at the sample surface. In this case, all the processes triggered by the UV pulse take place only after the VIS pulse impinged on the glass plate. No effect due to the UV laser pulse is visible in the profile. If, however, the UV pulse hits the target some 100 fs before the VIS laser pulse a low intensity spot appears at the center where the UV beam is focused as shown in Fig. 3b. The intensity of the beam 3.6×10^{13} W/cm^2 is high enough to create a highly ionized plasma on the surface of the sample. The ionization occurs on a time scale comparable to the pulse duration of the UV pulse. During a few 100 fs no significant energy or material transport can occur, hence, a plasma having nearly solid-state density is formed. Once the electron density reaches the criti-

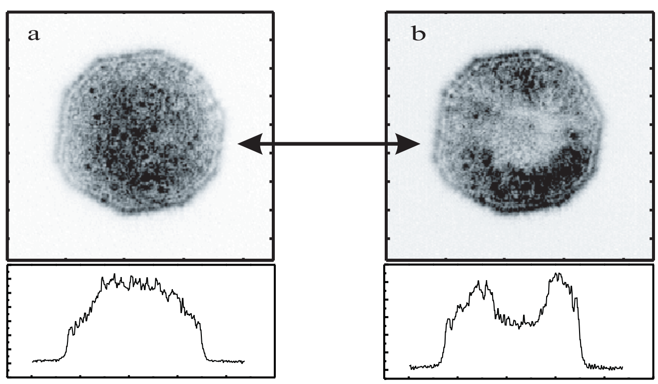

Fig. 3a,b. Images of the VIS laser pulse on the surface of the BK7 glass plate. **a** The delay time was adjusted so that the VIS pulse impinged before the UV laser pulse. **b** The UV pulse created a plasma on the surface of the BK7 glass plate before the VIS laser pulse hit the glass plate. Comparing the two horizontal intensity profiles clearly shows a low-intensity spot in the center of the green pulse which is due to plasma formation cal density for the VIS laser radiation $N_c = 4.45 \times 10^{21}\,\mathrm{cm}^{-3}$ the laser pulse is reflected resulting in a rapid decrease in transmission. The reflectivity of the plasma mirror reaches almost 100% and decays with a time constant of a few 10 to 15 ps [5]. After overlapping both foci spatially, the CCD camera was replaced by a photodiode equipped with a pinhole. The size and the position of the pinhole were chosen so that only the part of the VIS laser pulse that is affected by the UV laser pulse was transmitted (see Fig. 3b).

2 Results and discussion

In order to understand the transient behavior of the plasma mirror the following simple model was introduced. As has been shown in [6] the optical properties of dense plasmas can be well described by using the Drude model. Thus, assuming that the electron–ion collision rate is negligible compared to the light frequency and that the magnetic permeability of the bulk material is unity, the complex refractive index of the plasma can be written as

$$\tilde{n} = \sqrt{n_0^2 - \frac{\omega_p^2}{\omega_0^2}} \quad , \tag{1}$$

where n_0 is the (real) refractive index of the bulk material and ω_0 is the center frequency of the probe light. ω_p is the plasma frequency given as

$$\omega_p^2 = \frac{N_e e^2}{\varepsilon_0 m} \quad , \tag{2}$$

where N_e is the electron density in the plasma, e and m are the electron charge and mass, respectively. To calculate the instantaneous electron density we assume that it is proportional to the absorbed UV energy. (This means physically that we assume that there are no nonlinear processes in the plasma that lead to ionization which is, of course, not true in general but it can be used as a first-order approximation.) Then N_e can be obtained as

$$N_e(t) = \int_{-\infty}^{t} \mathrm{d}t' \, \frac{A}{d} \, I_{UV}(t') \quad , \tag{3}$$

where $I_{UV}(t')$ is the UV intensity, d the absorption depth (BK7: $d \approx 50$ nm), and A is a proportionality factor. The reflectivity of the surface can simply be calculated by using Fresnel's formula for perpendicular incidence

$$R(t) = \left| \frac{\tilde{n} - 1}{\tilde{n} + 1} \right|^2 \quad . \tag{4}$$

Finally, we obtain for the transmitted (VIS) energy at a time delay τ

$$T(\tau) = T_s \frac{E_{VIS} - \int_{-\infty}^{\infty} \mathrm{d}t' \, R(t') \, I_{VIS}(t' - \tau)}{E_{VIS}} \tag{5}$$

where T_s is the small signal transmission of the sample (at the VIS wavelength), and $E_{VIS} = \int_{-\infty}^{\infty} \mathrm{d}t' \, I_{VIS}(t')$. We want to stress that provided that the pulse duration of both pulses are known the above model contains only one fit parameter, i.e. the proportionality factor A.

The transmitted energy as a function of the delay is presented in Fig. 4. The squares represent the measured points while the solid lines were calculated by using (5). The data evaluation was accomplished in two steps. First the propor-

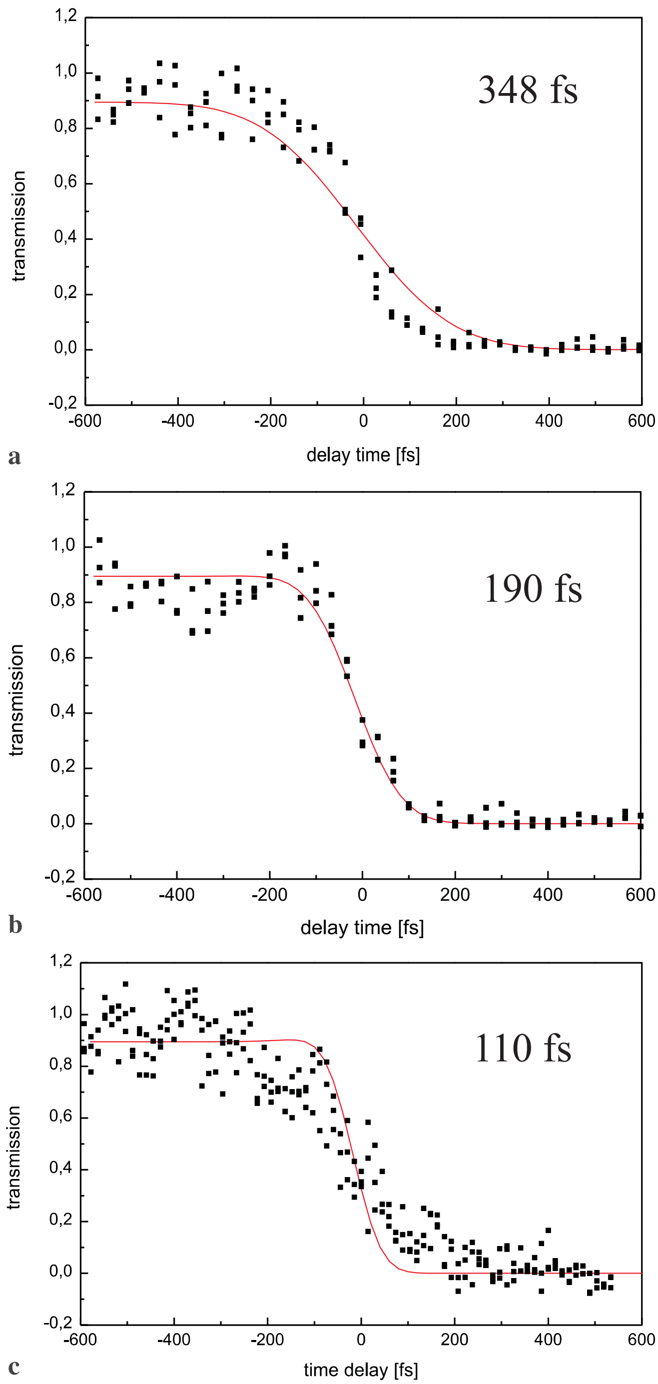

Fig. 4a–c. Cross-correlation measurements using the plasma shutter technique for different pulse width at 497 nm, 348 fs (**a**), 190 fs (**b**), and 110 fs (**c**), respectively. The width of the UV pulses was (215 ± 10) fs in all cases. The transmitted signal as a function of the delay time between the UV and the VIS laser pulse is shown. For negative delay times the pulse at 497 nm precedes the pulse at 248 nm and the measured transmission is one. The *solid lines* are calculated using the model described in the text

tionality factor was obtained from the best fit to the data given in Fig. 4b. This set of data was taken by moving the compressor to a position where the duration of the VIS laser pulse was approximately two times longer than the minimum belonging to optimum compressor setting. Thus, the measured pulse durations used as input parameters for the fit were (215 ± 15) fs for the UV and (190 ± 15) fs for the VIS laser pulse, respectively. The electron density was found to be $\geq 5 \times 10^{21}\ \mathrm{cm}^{-3}$ corresponding to a proportionality factor of $A = 627\ \mathrm{J}^{-1}$. For the next two measurements the compressor was changed resulting in a VIS pulse duration of (348 ± 15) fs and (110 ± 15) fs, respectively. The corresponding data are presented in Fig. 4a,c. The solid line represents the fit obtained by using the measured pulse duration and $A = 627\ \mathrm{J}^{-1}$. The obvious agreement between the data points and the solid indicates that the model presented above works reasonably well, considering the crude assumption of a linear ionization process. By making fits to the measured data based on pulse durations ranging from 110 fs to 600 fs we found that the residual error of the fit showed a slight tendency to slow down the rising edge which is fully consistent with the fact that the nonlinear processes in the plasma that are neglected in the model are expected to make the plasma build-up faster.

In any case, the build-up time of the critical density is less than the pulse width of the plasma creating pulse and it was found that the transmission of the delayed pulse increases again with a typical time constant of 10 to 15 ps. For the geometry shown in Fig. 1 the rapid increase of the electron density to values greater than the critical density corresponding to the VIS laser pulse serves as a ‘temporal knife edge’. As shown before, the resulting gate function may be used to measure the pulse width of the second laser pulse. In addition, it may be used as a powerful tool to find the temporal overlap between two pulses having different wavelengths. Since the lifetime of the plasma is on the order of a few 10 ps the data of a single experiment show which of the two pulses precedes the other. The only requirement is that one of the two pulses must have a minimum amount of energy in order to be able to create a plasma.

3 Conclusion

We have shown that the high-density plasma created by a UV pulse with a FWHM of about (215 ± 15) fs serves as an extremely fast switch for measuring the temporal intensity of a second short laser pulse with a different wavelength. In addition it provides a very simple method to overlap two fs laser pulses in time for subsequent pump-probe experiments.

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