Spatiotemporal characterization of VUV pulses by plasma-mirror frequency-resolve optical gating

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High-order harmonics of femtosecond laser pulses are known to be a powerful tool for time-resolved photoelectron spectroscopy and transient absorption spectroscopy of atoms and molecules. Short-wavelength pulses generated as high-order harmonics are also sensitive probes into laser plasma with a high density of excited electrons on the surface of solid materials, because the plasma resonance frequency is proportional to the square root of the density of excited electrons. Recently, we demonstrated that time-resolved vacuum ultraviolet (VUV) reflection spectroscopy is applicable to the characterization of VUV pulses and to the extraction of reflectivity changes in ultrafast plasma formation [1, 2]. In this study, we carefully investigate not only the temporal characteristic, but also the spatial mode of plasma reflection.

The output pulses of a Ti:Sapphire laser (80 fs, 795 nm, 10 Hz) are focused on a Xe gas jet for high-order harmonic generation. The fundamental (ω) and fifth harmonics (5ω) are separated with a multilayer mirror for 160-nm reflection. After adding a delay between ω and 5ω, two beams are colinearly recombined and focused on a transparent solid target of fused silica (FS). The target is continuously moved with a two-dimensional motorized stage such that the fresh surface is irradiated with laser pulses. Time-resolved VUV reflection spectra of FS are measured with a Seya–Namioka-type VUV spectrometer equipped with an imaging detector at the exit.

In the analysis, we improve the FROG iteration procedure based on the least-squares generalized projections algorithm [2]. As a result, the Fresnel reflection of the unexcited FS surface is included in the time-dependent reflectivity. The retrieved reflectivity is normalized by setting the reflectivity of the unexcited FS to unity. The spectrum component only by the ω irradiation is subtracted from the spectrum measured by the irradiation of both ω and 5ω.

The two-dimensional (2D) image at the exit of the spectrometer is recorded by a CCD camera in shot-by-shot mode. A raw single-shot image on the phosphor screen is shown in Fig. 1, where the horizontal axis is transformed to the wavelength and the vertical axis represents the spatial coordinate of the VUV beam in the vertical direction on the screen.

From the central part ($175 \le y \le 190$ pixels) in the vertical distribution, time-resolved VUV reflection spectra are obtained as shown in Fig. 2(a). In the negative-delay region, the weak Fresnel reflection of the unexcited FS surface is observed. Around zero delay ($\tau = 0$), the plasma is formed by the pump ω pulse and then the reflectivity rapidly increases.

Through the FROG analysis based on the LSGPA, we can successfully retrieve the time-resolved reflection spectra as shown in Fig.2(b). The 5ω pulse shape and temporal reflectivity are simultaneously characterized as shown in Fig. 3(a) and 3(b), respectively. The pulse duration of the 5ω pulse is extracted as 20 fs, which is nearly the Fourier-transform limit. The reflectivity of FS increases to the maximum R_{max} from the Fresnel reflectivity R_0 within the ω pulse duration of 80 fs. Thus, it is demonstrated that the 20-fs duration of the 5ω pulse can be characterized by PM-FROG, even if the rise time of the plasma reflectivity is longer than the 5ω pulse duration.

Next, we examine the spatial dependence of the 5ω

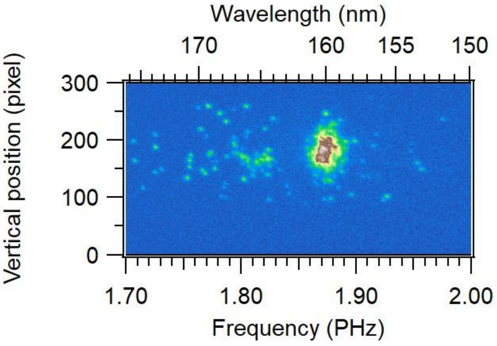

Fig. 1 Raw image of reflected VUV beam at the exit of VUV spectrometer.

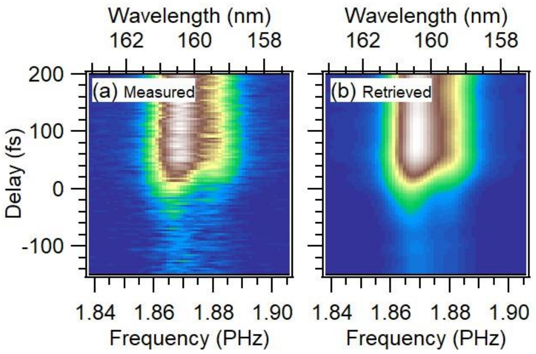

Fig. 2 Time-resolved reflection spectra of plasma mirror on FS. (a) Measured and (b) retrieved results.

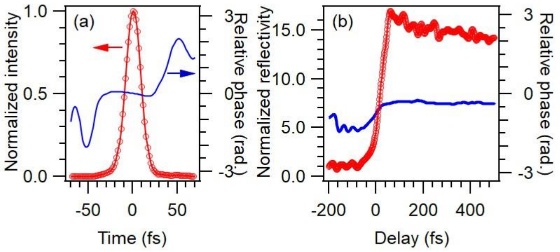

Fig. 3 (a) Retrieved 5ω pulse-shape. (Red circles: intensity, blue line: relative phase). (b) Time-dependent reflectivity (red circles) and its phase (blue line) of the plasma mirror. Reflectivity is normalized with the Fresnel reflectivity R_0 of the unexcited FS surface.

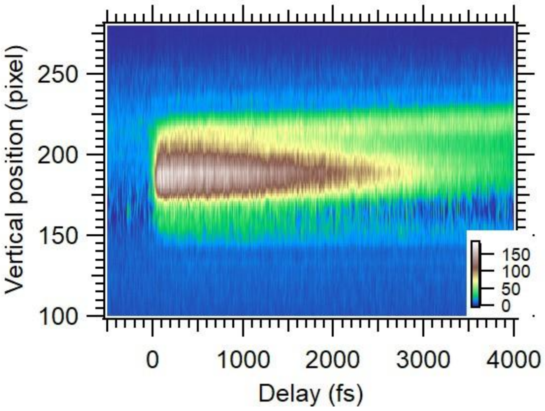

Fig. 4 Time-resolved spatial (vertical) profile of the reflected 5ω beam. The signal caused solely by pump-laser irradiation is subtracted.

waveform. The vertical spatial profile of the 5ω beam is obtained by integrating over the wavelength range between 159.1 and 161.2 nm. The temporal variation of the spatial profile is shown in Fig. 4. It is clear that there are two types of temporal behavior in the reflectivity. After the rapid increase, the reflectivity decreases almost to the initial value R_0 at the delay of $\tau = 4$ ps in the spatial region below y = 210 pixels. In the region above y = 210 pixels, a singularly slow decay of the reflectivity can be observed. It is known that a Seya–Namioka-type VUV spectrometer has inherent astigmatism. In other words, a high spectral resolution is achieved at the sacrifice of the vertical focusing. Nevertheless, a strong spatial dependence of the time-resolved reflectivity appears in the present measurement. It is worth discussing the spatial property of the plasma reflectivity qualitatively.

The maximum reflectivity $R_{\max}$ depends on the spatial region. This is attributed to the excited electron density governed by the spatial intensity profile of the pump ω beam, which should have the highest intensity in the spatially central part. As a result, the spatial distribution of the excited electron density exhibits the peak in the central region of the beam. The dependence of the reflectivity on the distance from the center reflects the spatial distribution of the excited electron density, as previously reported by Siegel et al. [3]. Although the reflectivity strongly depends on the spatial region, the VUV waveform is almost independent of the spatial region. In other words, the waveform of the VUV pulse is measured to be spatially uniform.

In addition to the major component, the singularly long decay component is also identified in the upper edge region of 210–230 pixels of the time-resolved spatial profile. This long decay is attributed to the reflection at the outermost ring of the ablation crater, where the fused silica surface survives after the ablation [3]. Although photoexcitation takes place at the outermost ring, the removal of the atomic layer hardly occurs. A certain amount of the atomic density is maintained at the outermost ring after the laser pulse irradiation, whereas the spallation takes place in the spatially central region. The decay rate in the upper edge region is expected to be governed dominantly by the electronic relaxation in the absence of ablation.

Further, it is found that the long decay component shifts spatially upward in the vertical direction as the delay increases. This shift reflects the dynamics occurring at the outermost ring. Material densification was proposed as a possible mechanism [3]. The spatial shift measured in this study is an indication of the radial expansion of the densification ring.

In summary, we demonstrated that a nearly Fourier-transform-limited VUV pulse with a pulse duration of 20 fs can be characterized by PM-FROG. In addition, the temporal waveform of the VUV pulse is measured to be spatially uniform, whereas the reflectivity depends on the spatial profile of the plasma mirror [4]. The spatial characterization of the VUV pulse becomes more important because of the rapid progress of ultrafast microscopy and coherent diffractive imaging in the short-wavelength region. Simultaneously, the spatiotemporal dependence of the reflectivity is also obtained by PM-FROG. The measurement technique of PM-FROG will be able to make a substantial contribution to ultrafast imaging in the VUV region.

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