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Time-resolved single-shot imaging of femtosecond laser induced filaments using supercontinuum and optical polarigraphy

Lihe Yan (闫理贺),[1,2] Xiaofang Wang (王小芳),[1] Jinhai Si (司金海),[1,a)] Shigeki Matsuo (松尾繁樹),[2] Tao Chen (陈涛),[1] Wenjiang Tan (谭文疆),[1] Feng Chen (陈烽),[1] and Xun Hou (侯洵)[1]

[1]*Key Laboratory for Physical Electronics and Devices of the Ministry of Education and Shaanxi Key Lab of Information Photonic Technique, School of Electronics and information Engineering, Xi'an Jiaotong University, Xianing-xilu 28, Xi'an 710049, China*

[2]*Department of Ecosystem Engineering, The University of Tokushima, 2-1 Minamijosanjimacho, Tokushima 770-8506, Japan*

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We have developed an ultrafast time-resolved imaging technique for the propagation dynamics of ultrashort laser pulses in transparent media. This method utilizes the optical polarigraphy technique and a chirped supercontinuum as the probe light. The supercontinuum senses the instantaneous birefringence induced by the propagation of an intense pulse, and a polarigraphy image with different color distributions could be obtained. Using this method, we performed a space-time characterization of the filament induced by a femtosecond laser pulse in CS_2, indicating that this technique could be used for the single-shot imaging of pulse propagation dynamics. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3694051]

The propagation of powerful ultrafast laser pulses in transparent media can induce some self-modulating nonlinear effects, such as self-focusing, self-defocusing, self-phase modulation, etc.[1–4] To observe this propagation behavior, indirect techniques such as the schlieren method has been used.[5] However, this method cannot take an instantaneous image of the interaction of the laser pulse with the material because only the long-lived gradient in the refractive index can be measured after the excitation. In the past few years, people have developed a direct method of femtosecond time-resolved optical polarigraphy (FTOP) for observing the pulse propagation of a focused femtosecond laser.[6–8] This method uses the instantaneous birefringence induced by the strong electrical field of the pulse in gases or liquids. Through consecutive femtosecond snapshot images of intense femtosecond laser pulses propagating in the medium, ultrafast temporal changes in the two-dimensional spatial distribution of the optical pulse intensity can be observed. However, the traditional FTOP method is limited to obtain the whole map of a single pulse's propagation instantaneously, as a mount of snapshot images are needed to be stacked to display the whole image.[6] Because these images are taken using a serial of independent pulses, it cannot fulfill the single-shot measurements of the spatial distribution of the propagation of an intense light pulse. In 2002, Fujimoto *et al.* demonstrated a successive four-frame instantaneous observation of an intense femtosecond optical pulse propagating in air.[9] However, a quadruple-pulse generator installed in the optical path is needed in the experiments. This will largely increase the difficulty and complexity of the experiments.

Supercontinuum, which can be generated by powerful ultrashort laser pulses propagating in transparent media, has found many applications in ultrafast measurements due to its ultra-broadband components and ultrashort duration.[10–12] In 1980s, people have developed ultrafast supercontinuum pump-and-probe absorption technique to study the relaxation processes in solid-state physics, chemistry, and biological systems.[13] Combining with the optical Kerr gate (OKG) technique, Yasui and his coworkers have realized simultaneous three dimensional (3-D) imaging of objects using a chirped supercontinuum.[14] In 2007, Ye *et al.* reported a detection mechanism for ultra-broadband multicolor fluorescence detection using an ultrafast supercontinuum source to simultaneously excite different fluorophores.[15]

In this paper, we report an ultrafast time-resolved imaging technique for the propagation dynamics of ultrashort laser pulses in transparent media. This method utilizes the optical polarigraphy technique and a chirped supercontinuum as the probe light. The supercontinuum senses the instantaneous birefringence induced by the intense light pulse, and a polarigraphy image with different color distribution can be obtained. Using this technique, we perform a space-time characterization of the filament induced by femtosecond laser pulse in CS_2, indicating that this method is useful in the monitoring the propagation dynamics of an intense light pulse in a transparent medium.

Figure 1 shows the experimental setup for optical polarigraphy scheme. A Ti:sapphire amplifier system emits 30 fs laser pulses centered at 800 nm at a repetition rate of 1 kHz with horizontally linear polarization. The laser beam is split into a pump and a probe beam by a beam splitter. After passing through a delay line, the polarization of the pump beam is changed to vertical using a half-wave plate. The pump beam is focused into a 10-mm long fused silica cuvette filled with CS_2 by a 100 mm lens. For a pulse of 8 μJ energy in our experiments, the nonlinear focus was located at about 1 mm inside the input window of the cuvette.

The probe beam is focused into a 10-mm cell filled with distilled water to generate a supercontinuum by a 100-mm lens. To temporally broaden the supercontinuum pulse, a 20-mm thick fused silica is introduced into the optical path of

[a)]Author to whom correspondence should be addressed. Electronic mail: jinhaisi@mail.xjtu.edu.cn.

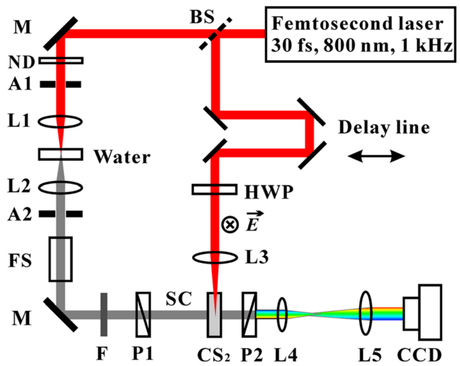

FIG. 1. (Color online) Experimental setup for optical polarigraphy, where a supercontinuum generated in water was used as the probe light. BS: beam splitter, M: mirror, ND: neutral density filter, A: aperture, L: lens, FS: fused silica, F: short-wave pass filter, P: polarizer, and HWP: half-wave plate.

the probe beam. The supercontinuum, after passing a short-wave-pass filter to remove the 800 nm light and the infrared part, is collimated and introduced into the CS_2 cell perpendicularly to the direction of the pump path. The light spot of the supercontinuum covers the area of the focal point of the pump beam. In front of the sample, a polarizer (P1) is set to 45° with respect to the horizontal plane of the optical stage and allows parts of the supercontinuum to pass. When the pulse passes through the interaction region, only the components perpendicular to the polarizer can be extracted by the analyzer (P2) placed behind the sample. To record the polarigraphy image, a high-spatial-resolution CCD camera is located on the imaging plane of the filaments.

Through nonlinear interactions including self-phase modulation, self-steepening, stimulated Raman scattering, and four-wave mixing,[10–12] a broad supercontinuum was produced extending down to 450 nm, as shown by the inset of Fig. 2. After passing through a short-wave-pass filter, the 800 nm light and the infrared part of the supercontinuum were removed. Figure 2 shows the temporal behavior of the supercontinuum generated in water measured using ultrafast OKG method.[16,17] The duration of the supercontinuum was estimated to be more than 20 ps, as the chirped pulse was broadened by the fused silica and optical elements in the setup due to the group velocity dispersion (GVD) effect.

Figure 3(a) shows the recorded polarigraphy image of the pump pulse propagation in CS_2. To increase the signal-to-noise ratio, the exposure time of the CCD camera is set at

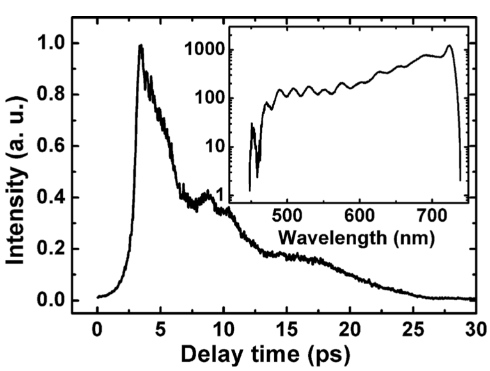

FIG. 2. Temporal behavior of the supercontinuum generated in water by a femtosecond laser pulse. The inset shows the spectrum of the supercontinuum after passing through a short-wave-pass filter.

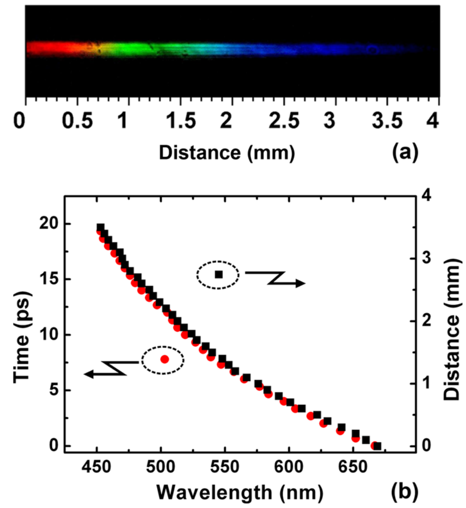

FIG. 3. (Color online) (a) Imaged filament induced by a femtosecond laser pulse in CS_2, in which 133 shots are integrated to improve the signal-to-noise ratio. (b) Chirp character of the supercontinuum (red circles) and wavelength distributions of the recorded image (solid squares).

1/6 s and 133 shots are integrated to produce the image. The pulse propagates from the left to right. Due to the chirp character of supercontinuum, different wavelength components overlapped with pump pulse at different propagating time in CS_2. The recorded image color changes from red to blue, while different color corresponds to different time of the pulse propagating in the medium. Because of the balance between Kerr self-focusing and plasma defocusing induced by the nonlinear ionization, a filament was produced in CS_2. The recorded filament lasts about 4 mm long in CS_2.

The red circles in Fig. 3(b) show the chirp character of the supercontinuum measured using OKG method. The left axis of the figure indicates the temporal distributions for different wavelength components, while the right axis shows the calculated propagation distance of an 800 nm pulse in CS_2 in the corresponding time. Here, the linear refractive index of 1.62 at 800 nm for CS_2 was used. Then, we measured the wavelength distributions of the probe light passing through the analyzer (P2). By finely moving a 200-μm core diameter fiber along the pulse propagation direction behind the analyzer (P2), a serial of spectra centered at different wavelength were coupled into a spectrometer. The solid squares in Fig. 3(b) indicate the spatial distributions of the spectra with different central wavelength, which accord well with the chirp character of the supercontinuum measured beforehand. Therefore, by selecting a wavelength at a certain distance from the recorded image, one can finely determine the propagation time of a pulse in an indeterminate medium, as well as the spatial intensity distribution at the corresponding time by referencing the chirp character of the supercontinuum.

Furthermore, as the supercontinuum shows a positive chirp, a narrow spectrum band will show impulsive property. Hence, one can determine the temporal intensity evolution of the pump pulse during its propagation by analyzing the exposure intensity evolution of different spectrum bands. An optional method is to use the Hue-Saturation-Value (HSV) color model to represent the recorded image. By analyzing the brightness of the map in a certain hue region, one can

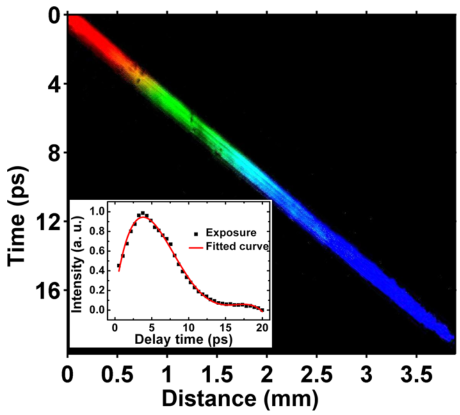

FIG. 4. (Color online) World line of the filament induced by a femtosecond laser pulse in CS_2. The inset shows the CCD exposure intensity for different wavelength components of the supercontinuum.

obtain the transient refractive index map of the pump pulse at the corresponding distance.

However, three main factors might influence the image intensity: the CCD response to different wavelength, the intensity distributions for different wavelength components in the supercontinuum, and the Kerr response to different wavelength components of the medium. This would distort the intensity distribution of the recorded polarigraphy images. Using a 0.1-mm slit placed behind the analyzer, we measured the exposure intensity for different wavelength components by moving the delay line. The results are shown by the squares in the inset of Fig. 4, in which the solid line indicates the fitted results using a high-order polynomial. Using this, the exposure intensity of the image was normalized, and the world line of the pump pulse propagating in CS_2 liquid was obtained as shown by Fig. 4. The vertical and horizontal axis corresponds to the time and distance of the laser pulse propagation, respectively. The pulse propagates from top-left to bottom-right.

In conclusion, utilizing the optical polarigraphy technique and a chirped supercontinuum, we have developed an ultrafast time-resolved imaging technique for the propagation dynamics of ultrashort laser pulses in transparent media. The supercontinuum senses the instantaneous birefringence induced by an intense light pulse, and a polarigraphy image with different color distribution could be obtained. Using this method, we performed a space-time characterization of the filaments induced by femtosecond laser pulse in CS_2, indicating that this imaging technique could be used for the ultrafast single-shot measurements of pulse propagation dynamics. By analyzing the recorded image using HSV model, one can obtain the transient refractive index map of the pump pulse at a certain propagating distance.

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