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Ultrafast single-shot imaging of femtosecond pulse propagation in transparent liquids using a supercontinuum and optical polarigraphy

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Abstract
We investigated the ultrafast time-resolved single-shot imaging of femtosecond pulse propagation using a supercontinuum and optical polarigraphy in transparent liquids. The probe supercontinuum senses the instantaneous birefringence induced by the laser pulse, and a polarigraphy image with different color distributions could be obtained. By comparing the wavelength distributions and the saturation variation of the images, the recorded polarigraphy images in two samples with different response time were analyzed. In the fast response sample, *N*-methyl-2-pyrrolidone (NMP), the spectral widths and the saturation values of the polarigraphy image at fixed positions were narrower and higher than those in CS_2. Due to the slow response of CS_2, the probe light sensed a long-lived birefringence and the polarigraphy image contained more wavelength components at every position along the pump pulse propagation direction.

Keywords: single-shot imaging, femtosecond time-resolved, optical polarigraphy, supercontinuum

(Some figures may appear in colour only in the online journal)

1. Introduction

The advent of femtosecond laser necessitates research of the propagation characteristic of ultrashort and intense laser pulses, which involves various self-modulating nonlinear effects [1, 2]. The propagation mode of laser filamentation has aroused great interest due to its numerous potential applications, such as atmospheric analysis, power supply for high-speed electric vehicles, and generation of terahertz radiation [3–5]. 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, based on the optical Kerr effect, uses the instantaneous birefringence induced by the strong electrical field of the pulse in transparent materials. 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 can hardly obtain the whole map of a single pulse's propagation instantaneously, as a collection of snapshot images needs to be stacked to display the whole image [6]. Because these images are taken using a series of independent pulses, the technique cannot fulfil the single-shot measurements of the spatial propagation distribution of an intense light pulse. And this single-shot technique is especially important for research into the irreversible effect

of an intense pulse with the propagation medium. 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 greatly increase the difficulty and complexity of the experiments.

In our previous report, we demonstrated an ultrafast single-shot imaging method utilizing the optical polarigraphy technique and a chirped supercontinuum as the probe light. The supercontinuum senses the instantaneous birefringence induced by the intense light pulse in transparent material, and a polarigraphy image with different color distribution is obtained, allowing the imaging of the propagation dynamics of a single laser pulse [10]. However, the Kerr medium of CS_2 is of slow time response; thus, in the femtosecond regime, the probe light would sense residual birefringence, thereby reducing the temporal resolution of the polarigraphy image.

In this study, we performed the single-shot imaging of femtosecond laser pulses propagating in *N*-methyl-2-pyrrolidone (NMP) and CS_2, respectively, using the polarigraphy technique and a supercontinuum. Because of the chirp character of the supercontinuum, the wavelength distributions of the polarigraphy images can agree well with the pulse propagation time. The spectral widths at fixed positions in CS_2 are broader than in NMP due to its slow time response. Though image analysis using the hue–saturation–value (HSV) model, it is found that the saturation in CS_2 is lower than in NMP on account of mixing of more color component at every corresponding position.

2. Experimental methods

In our experiments, we selected two transparent liquids, NMP and CS_2, as the Kerr media. First, the nonlinear responses of the two samples were measured using the time-resolved optical Kerr gate (OKG) method. It was performed using 65 fs, 800 nm pulses from a regeneratively amplified Ti: sapphire laser system (Libra-USP-HE, Coherent Inc.) operating at 1 kHz repetition rate with horizontally linear polarization. The measurement setup used was similar to that reported elsewhere [11]. Figure 1 shows the time-resolved OKG signals of the samples. The red circles and the black squares in figure 1 refer to the OKG signals of NMP and CS_2, respectively. The red solid curve indicates the Gaussian fit of OKG signals of NMP, the full-width at half-maximum (FWHM) of which is about 120 fs. As the OKG singles for NMP show no obvious slow decay process, the origin of the nonlinear response of NMP could be mainly attributed to electronic process, the characteristic time of which is much faster than 120 fs. The black curve in figure 1 indicates the decay process of the nonlinear response of CS_2, which is fitted using a bi-exponential decay function with time constants 160 fs and 1.6 ps. The fitted result agrees well with those reported in the previous research, indicating that the slow decay process of the nonlinear response in CS_2 should be mainly attributed to molecular orientation relaxation [12–14].

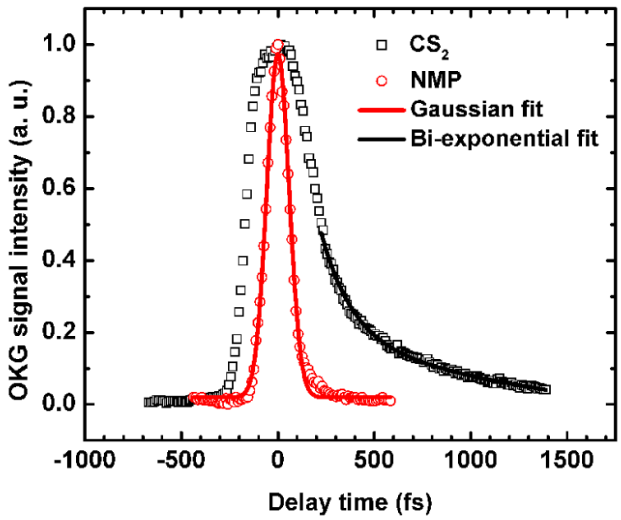

Figure 1. Time-resolved measurements of OKG signals for NMP and CS_2. The red circles and the black squares are the experimental data of NMP and CS_2, respectively. The red solid curve indicates the Gaussian fit of NMP data. The black curve for CS_2 data is fitted by a bi-exponential decay function with time constants 160 fs and 1.6 ps.

Then, we performed the single-shot imaging of femtosecond laser pulses propagating in NMP and CS_2 using the polarigraphy technique and a supercontinuum. The general scheme of the experiment is shown in figure 2. The laser beam was split into a pump and a probe beam by a beam splitter. After passing through a delay line, the polarization of the pump beam was changed to vertical using a half-wave plate. The pump beam was focused into a 10 mm long fused silica cuvette filled with the sample (NMP or CS_2) by a 100 mm lens. The nonlinear focus of the pump beam was located at about 1 mm inside the input window of the cuvette. The probe beam was focused into a 10 mm cell filled with distilled water to generate a supercontinuum by a 100 mm lens. Through nonlinear interactions including self-phase modulation, self-steepening, stimulated Raman scattering, and four-wave mixing, the broad supercontinuum was produced extending down to 450 nm [15–18]. To temporally broaden the supercontinuum pulse, a 20 mm thick fused silica was introduced into the optical path of the probe beam. The supercontinuum, after passing a short-wave-pass filter to remove the 800 nm light and the infrared part, was collimated and introduced into the sample cell perpendicularly to the direction of the pump path. The light spot of the supercontinuum covered the area of the focal point of the pump beam. In front of the sample, a polarizer (P1) was set to 45° with respect to the horizontal plane of the optical stage, allowing parts of the supercontinuum to pass. When the pulse passed through the interaction region, only the components perpendicular to the polarizer could be extracted by the analyzer (P2) placed behind the sample. To record the polarigraphy image, a high-spatial-resolution CCD camera was located on the imaging plane of the filaments.

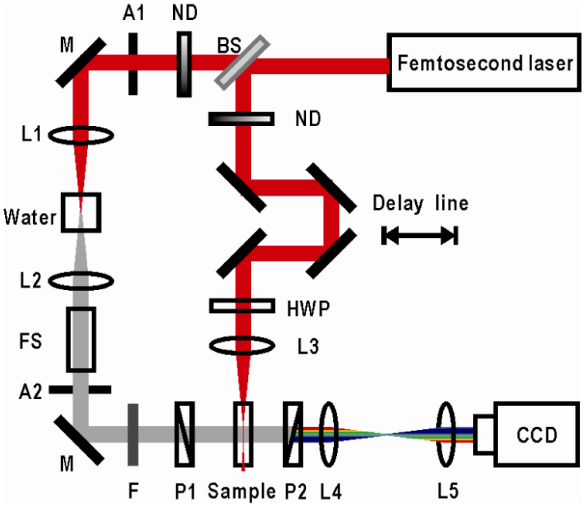

Figure 2. Experimental setup for optical polarigraphy, where a supercontinuum generated in water was used as the probe light. BS: beam splitter, M: mirror, ND: neutral density filter, A: aperture, L: lens, FS: fused silica, F: short-wave-pass filter, P: polarizer, and HWP: half-wave plate.

3. Results and discussion

The recorded FTOP images of the pump pulse propagation in NMP and CS_2 are shown in figures 3(a) and (b), respectively. To increase the signal-to-noise ratio, the pump powers were fixed at 44.5 mW in NMP and 11.5 mW in CS_2, and the exposure times during the measurements in the samples were set to be 1/6 s in NMP and 1/75 s in CS_2. The laser pulse traveled from left to right. Due to the chirp character of the supercontinuum, different wavelength components overlapped with the pump pulse at different propagating times in the samples. The colors of the recorded images change from red to blue: 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 a sample. The recorded filaments last about 3 mm long in samples.

Then, we analyzed the relation between the chirp character of the supercontinuum and the spatial spectra distribution of FTOP images. The chirp character of the supercontinuum was measured using the OKG method [19]. The red circles in figures 3(c) and (d) indicate the temporal distributions for different wavelength components of the supercontinuum. The spatial spectra distributions of the FTOP images were obtained by finely moving the fiber along the pulse propagation direction behind the analyzer (P2). For comparison, we converted the propagation distance of the 800 nm pulse to the propagation time in the samples. Here, the linear refractive index values 1.62 for CS_2 and 1.47 for NMP at 800 nm were used. In figure 3, the black squares indicate the spatial spectra distributions of the FTOP images, 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 accurately 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, we compared the difference of the FTOP images recorded in NMP and CS_2, which showed fast and slow responses, respectively. From figures 3(a) and (b) we can see that the color of the recorded FTOP image in NMP is purer than that in CS_2. To explain this phenomenon, we measured the spectra of the FTOP images at different positions by moving the fiber optic spectrometer behind the analyzer (P2)

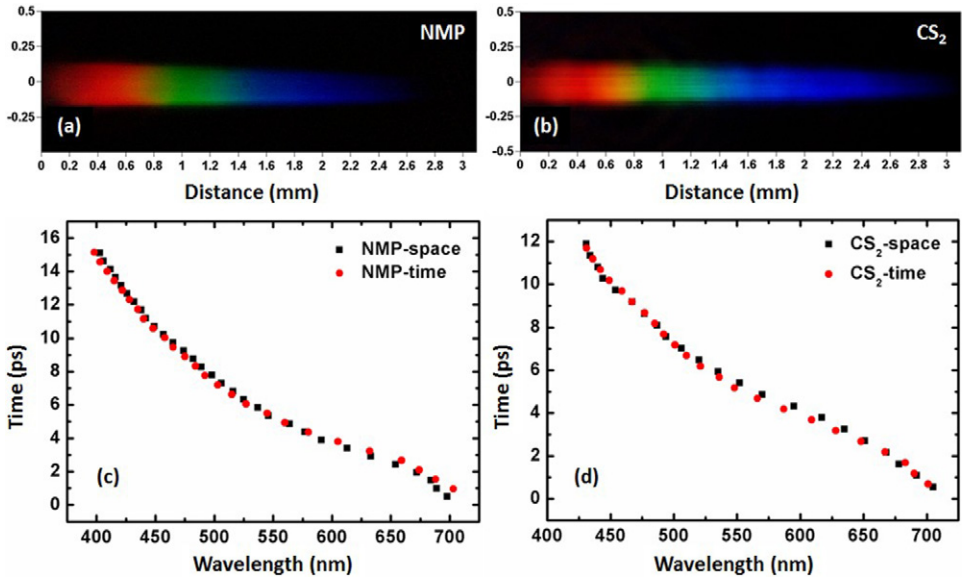

Figure 3. Imaged filaments induced by a femtosecond laser pulse in (a) NMP and (b) CS_2; the CCD exposure times are 1/6 s and 1/75 s, respectively. Distance means the propagation distance of the pump pulse in the irradiated range of the supercontinuum. Chirp character of the supercontinuum (red circles) and wavelength distributions of the recorded images (black squares) in (c) NMP and (d) CS_2.

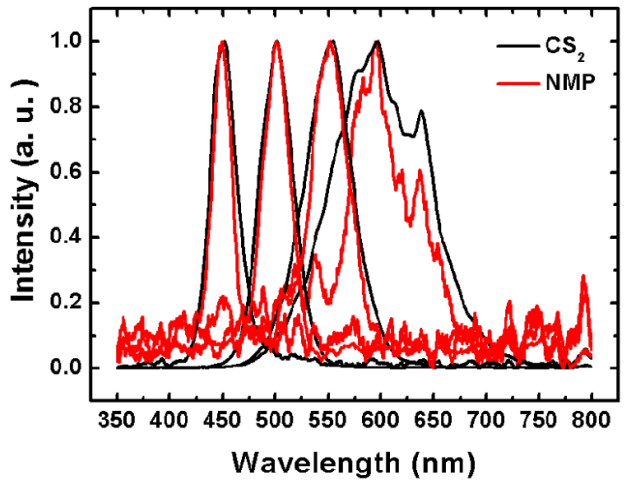

Figure 4. The spectra of the FTOP images along the pulse propagation direction detected by the fiber optic spectrometer behind the analyzer (P2).

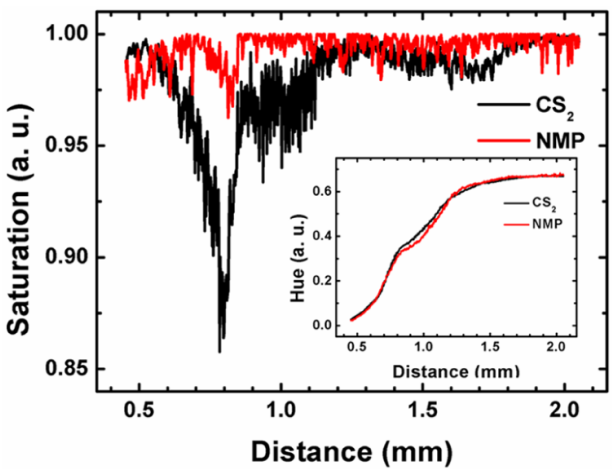

Figure 5. The saturation of the FTOP images along the pulse propagation direction. The inset shows the corresponding hue of the images.

along the pulse propagation direction. The fiber diameter is about 100 μm. In figure 4, the red line and the black line indicate the spectral lines of NMP and CS_2, respectively, where the central wavelengths were fixed at 450 nm, 500 nm, 550 nm, and 600 nm. Because CS_2 has a much longer response time than NMP, a residual birefringence would be experienced by the subsequently arriving spectral components of the probe supercontinuum. Hence the recorded spectra in the detecting region should be wider in CS_2 than those in NMP. In our experiments, however, the spectra of the FTOP signal were coupled into the spectroscopy using a fiber with a 100 μm diameter. In the collecting region of 100 μm, the propagation time of the pump pulse was estimated to be about 500 fs in the samples, which was much longer than the response time of NMP and comparable to the FWHM of the OKG signals of CS_2. Hence, the spectra difference was not as remarkable as the difference of the response times of the two samples.

For further understanding, we used the HSV color model to analyze the recorded images, as shown in figure 5. HSV stands for hue, saturation, and value. Hue scales the perceived colors: red, yellow, green, and blue, or a combination of two of them. Saturation is a ratio of chroma to the maximum chroma of a color, which indicates the chromatic purity. Value, also often called brightness, is defined as the location of a visual perception along a continuum from black to white. In figure 5, the red line and the black line show the saturation curves of the polarigraphy images in NMP and CS_2, respectively. The horizontal axis indicates the propagation distance of the laser pulse, which is consistent with that in figure 3. The corresponding hue curves of the images are presented in the inset of figure 5; they show almost the same variation tendency. The saturation of the FTOP image in NMP is higher than that in CS_2, indicating that the color purity of the polarigraphy image in NMP is much higher. Due to the slow response of CS_2, a residual birefringence would be left when the pump pulse passed through a fixed position in the sample. Thus, more wavelength components of the probe light would be affected and would pass through the analyzer, resulting in the chromatic impurity of the FTOP image. In addition, the colors of the FTOP images change rapidly over the propagation distance 0.5–1.2 mm, as indicated by the hue curves shown in the inset of figure 5. Therefore, different wavelength components were mixed more easily in the FTOP image of CS_2 due to its slow response, and a vast gap in the vicinity of 0.8 mm between the saturation curves was observed.

4. Conclusions

In conclusion, we have investigated the ultrafast time-resolved single-shot imaging of femtosecond filaments using a supercontinuum and the FTOP method in NMP and CS_2. The wavelength distributions of the polarigraphy images corresponded with the pulse propagation time because of the chirp character of the supercontinuum. In the slow time response sample CS_2, the spectral widths at fixed positions were broader than those in the fast time response sample NMP. This was because, in the slow response sample, the probe light experienced the residual birefringence of the pump light. Thus, the FTOP image contained more wavelength components at every position along the pump pulse propagation direction in the slow time response sample, and the FTOP image in the fast time response sample had higher saturation values.

Acknowledgments

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