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**Abstract.** Using a femtosecond time-resolved optical polarigraphy (FTOP) imaging technique, we measured the ultrafast propagation dynamics of femtosecond laser pulses in transparent materials, $CS_2$ and fused silica, respectively. The FTOP images showed different profiles in these two media due to their different nonlinear response time. Based on the FTOP technique, a femtosecond time-resolved single-shot optical Kerr effect measurement was demonstrated, which can be accomplished using a single-laser shot and has a time resolution of about 100 fs. The polarization dependence of the image intensity indicated that the FTOP images were mainly induced by the transient birefringence effect induced by the pump pulse.  [DOI: 10.1117/1.OE.53.5.056101]

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## 1 Introduction

For the last few decades, the propagation of powerful ultrafast laser pulses has attracted much interest owing to its self-modulating nonlinear effect, such as self-focusing, self-defocusing, self-phase modulation, etc.[1–4] In recent reports, the propagation of femtosecond intense laser pulses in an underdense plasma has also created much interest because of its importance in laser fusion, particle acceleration, and high-field physics in laser–plasma interaction.[5–7] To observe this propagation behavior, indirect techniques such as the schlieren method has been used.[8] However, this method could not take an instantaneous image, because one could observe only the plasma profile that had a long decay time after the excitation. Therefore, even when a femtosecond laser was used as the excitation source, only the phenomena on the time scale affected by the long decay of the plasma could be observed.

To overcome these shortcomings, people have developed a new method for visualizing directly the intensity distribution of a light pulse in a femtosecond time regime.[9] The method, femtosecond time-resolved optical polarigraphy (FTOP), makes use of Kerr effect induced by electric field of the laser. Because the induced birefringence has an ultrafast response in gases or liquids, an instantaneous image of the laser pulse in the interaction region is able to be recorded. By using FTOP imaging technique, we were able to observe the ultrafast temporal dynamics of the propagation of intense femtosecond optical pulses.

On the other hand, because the intensity of the FTOP image is proportional to the induced birefringence inside the medium, the nonlinear response, i.e., the response time and the nonlinearity of the sample can be determined from the FTOP image. As the FTOP image can be recorded using a single-laser pulse, the nonlinear response of the sample can be measured using a single-laser shot. Compared with the traditional optical Kerr measurements via multiple-shot experiments, this method can avoid the influence of the irreproducibility of the laser pulses and the irreversible change of the sample caused by the intense laser pulse.

In this article, 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 400-nm beam as the probe light. Using this technique, we measured the ultrafast propagation dynamics of femtosecond laser pulses in transparent materials, $CS_2$, and fused silica, respectively. Through consecutive snap-shot imaging of the laser pulses propagating in the media, ultrafast temporal changes in the spatial distribution of the optical pulse intensity were obtained. The FTOP images showed different profiles in these two media due to their different nonlinear response time. Based on the FTOP technique, a femtosecond time-resolved single-shot optical Kerr effect (OKE) measurement was demonstrated, which can be accomplished using a single-laser shot and has a femtosecond time resolution. The polarization dependence of the image intensity indicated that the FTOP images were mainly induced by the transient birefringence effect induced by the pump pulse.

## 2 Experiment

Figure 1 illustrates the experimental setup. A Ti:sapphire amplifier system (Libra-USP-HE, Coherent Inc., Santa Clara) emits 65-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 beams by a beam splitter. After passing through a delay line, the polarization of the pump beam is changed to vertical

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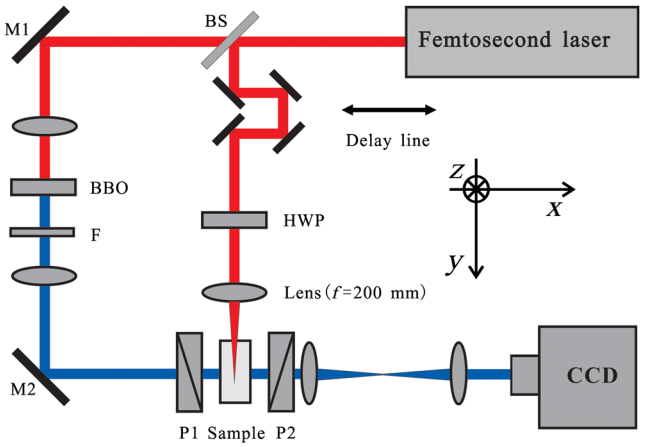


**Fig. 1** Experimental setup for optical polarigraphy. BS: beam splitter, M: mirror, L: lens, P: polarizer, and HWP: half-wave plate, F: bandpass filter.

using a half-wave plate. The pump beam is focused into a 10-mm-long fused silica cuvette filled with the liquid sample by a 200-mm lens. For a pulse of 8-$\mu$J energy in our experiments, the nonlinear focus was located at about 1 mm inside the input window of the cuvette, and no optical damage was observed under these conditions.

The probe beam is frequency doubled to 400 nm by a $\beta$-barium borate (BBO) crystal. After passing a bandpass filter centered at 400 nm, the probe beam is collimated and introduced into the sample cell perpendicularly to the direction of the pump path. The light spot of the probe beam covers the area of the focal point of the pump beam. In front of the sample, a polarizer (P1) is set to 45 deg with respect to the horizontal plane of the optical stage and allows parts of the probe beam 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 FTOP image, a high-spatial resolution CCD camera (Digital Camera DXM1200F, Nikon, Tokyo, Japan) with a $4f$ system is located on the imaging plane of the filaments. The spatial resolution of the imaging system is 3.4 $\mu$m/pixel.

## 3 Results and Discussion

First, we recorded the FTOP images of the pump pulse propagating in $CS_2$ and fused silica at different time by changing the optical delay of the probe pulse. Figures 2(a)–2(d) and Figs. 2(e)–2(h) show the typical FTOP images of pump pulse propagating at different moments in $CS_2$ and fused silica, respectively. The pump power was 10 mW and propagated from left to right, and time proceeds from top to bottom. To increase the signal to noise ratio, the exposure time of the CCD camera for each image was set at $1/2$ s, corresponding to the total pulse number of 500, and the background such as the emission from the breakdown plasma was subtracted. The brightness of images was normalized and the correspondence between the brightness and the image color is given at the bottom of the figures. From the figure, we can see that the transverse size of the spots in each sample remains the same as the pump pulse propagating, that is because of the balance between Kerr self-focusing and plasma defocusing induced by the nonlinear ionization.

In order to interpret the propagation behaviors of femtosecond pulses in the media clearly, we illustrate snap-shot images in the two samples at 1 ps. Figures 3(a) and 3(b) show the polarigraphy image in fused silica and $CS_2$, respectively. From the figure, we can see that the FTOP image in $CS_2$ has an asymmetrical decay tail, while that in fused silica showed a symmetrical distribution. This could be attributed to the different nonlinear response the two samples. As the nonlinearity of $CS_2$ originated from the orientation of the molecules has a long response time, the residual birefringence might cause the transmittance of the probe pulse even when the pump pulse has passed. Hence, the FTOP image showed an asymmetric profile when an ultrashort laser pulse propagated in $CS_2$.

The squares and circles in Fig. 3(c) show the normalized intensity distribution of FTOP images along the axial direction recorded in $CS_2$ and fused silica, respectively. The horizontal axis corresponds to the delay time between pump and probe pulses. In the FTOP measurements, the horizontal length of the image corresponds to the response time of the OKE. Hence, we fitted the intensity distribution of the FTOP images in both samples respectively. The red line in Fig. 3 indicates the nonlinear response of fused silica, which was fitted using a Gaussian equation. The full width at half maximum was estimated to be about 160 fs, which showed no slow response, indicating that the OKE signals in fused silica was mainly originated from electronic process.[10] For $CS_2$, the nonlinear response showed an obvious slow relaxation process. Using an exponential function, we fitted the decay processes of image intensity as

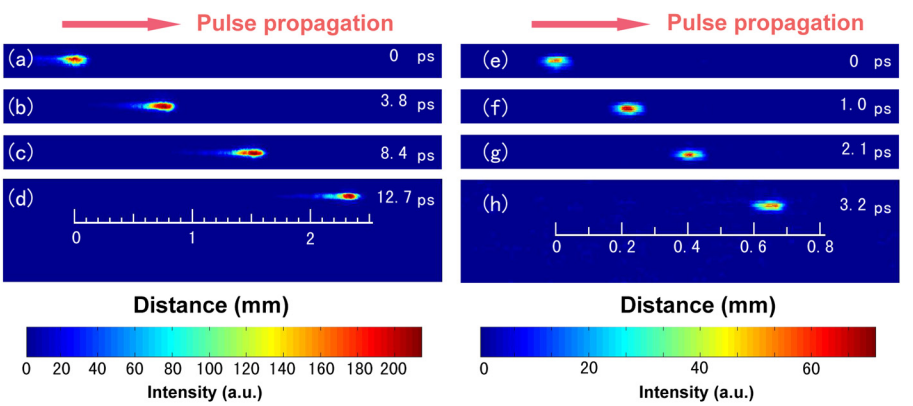


**Fig. 2** Images of a 10-mW pump pulse propagation in (a)–(d) $CS_2$ and (e)–(f) fused silica at different time.

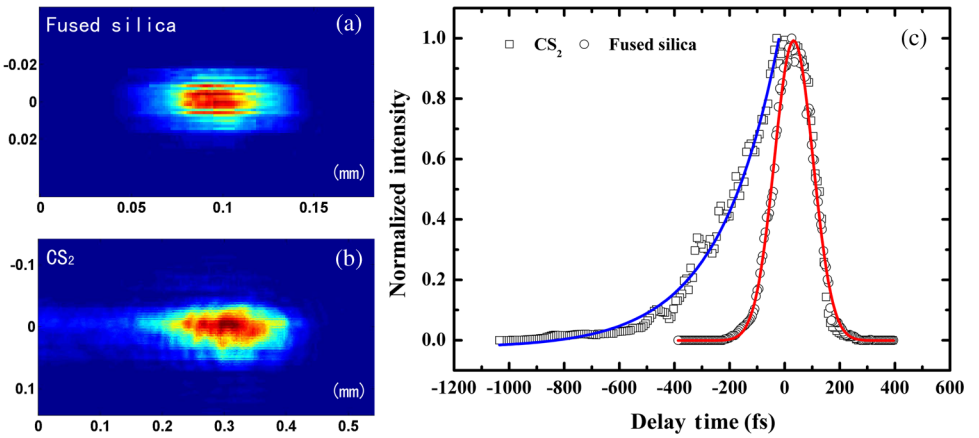


**Fig. 3** Single-shot measurements of the optical Kerr effect (OKE) response for $CS_2$ and fused Silica based on femtosecond time-resolved optical polarigraphy (FTOP) technique. (a) FTOP image in fused silica, (b) FTOP image in $CS_2$, and (c) time-resolved OKE response for $CS_2$ and fused silica.

given by the blue line in Fig. 3. The exponential decay time in $CS_2$ was estimated to be about 1.1 ps, which agreed well with the previous reports.[11] The origination of the nonlinear response in $CS_2$ was mainly attributed to the reorientation of molecules.

It should be noted that a femtosecond time-resolved single-shot measurements of the nonlinear response in materials can be accomplished using this method, as one can extract the OKE response of the media from the single-shot FTOP images. The time resolution of the single-shot OKE measurements is mainly limited by the pulse duration. Compared with the traditional femtosecond time-resolved OKE measurements, this method can be accomplished using a single-laser shot, while the former one need to record the OKE signals at different delay time separately via multiple laser shots.[12] Using this single-shot measurements technique, the nonlinear response for materials can be accurately acquired, while some irreversible change might be induced after multiple laser pulse irradiation.[13]

To better understand the origin of FTOP, we measured the dependence of the polarigraphy intensity on the polarization direction of the pump pulse. We assume that the linearly polarized pump and probe light propagated along the $x$ and $y$ axes, respectively, as shown in Fig. 1. When the pump light polarizes in the $yoz$ plane, it will induce a refractive index change on $x$ and $z$ axes, respectively. When the polarization angle of the probe light is fixed at 45 deg with respect to $z$ axis, and enters the interaction region, the probe beam senses different refractive indices between the $x$ and $z$ axes. Thus, a phase shift occurred between these two directions, which can be given by:

$$\Delta\phi = \phi_x - \phi_z = \frac{\omega}{c}\frac{3\omega}{kc}(\chi^{(3)}_{1111} - \chi^{(3)}_{1221})|E(\omega)\cos\theta|^2 z, \qquad (1)$$

where $\chi^{(3)}_{1111}$ and $\chi^{(3)}_{1221}$ are the tensor component of the effective third-order susceptibility. $E(\omega)$ is the strength of the pump electric field. $\theta$ corresponds to the angle between the polarization direction of the pump and $z$ axis. In the photoinduced birefringence effect, the intensity of the probe beam that passed through the analyzer is given as[13]

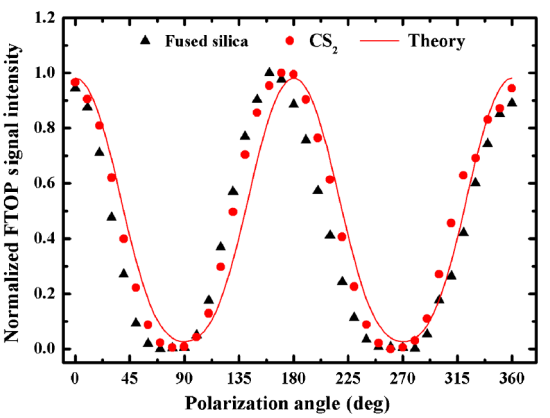


**Fig. 4** Polarization dependence of FTOP signals. The rounds and triangles are experimental results. The red solid curve is calculated from Eq. (1) to fit the experimental data.

$$I_{\text{polarigraphy}} \propto \sin^2\left(\frac{\Delta\phi}{2}\right) = \sin^2\left[\frac{\omega}{c}\frac{3\omega}{kc}\frac{L}{2}(\chi^{(3)}_{1111} - \chi^{(3)}_{1221})|E(\omega')\cos\theta|^2\right]. \qquad (2)$$

Here, $L$ is the interaction length between pump and probe. The circles and triangles in Fig. 4 show the FTOP signal intensity as a function of the angle between the pump polarization direction and the $z$ axis in $CS_2$ and fused silica, respectively. The red solid curve shows the fitted results using Eq. (2). The polarization dependence of the FTOP signal intensity showed a period of $\pi$, with the maximum and minimum values occurring at $n\pi$ and $\pi/2 + n\pi$ $(n = 0, 1, 2, \ldots)$, respectively. We can find from the figure that the theoretical calculation data agrees well with the experimental results, indicating that the FTOP images were mainly originated from transient birefringence effect.

## 4 Conclusions

In conclusion, utilizing the optical polarigraphy technique, we achieved an ultrafast time-resolved imaging technique for the propagation dynamics of ultrashort laser pulses in

transparent media. Using this method, we clearly and directly observed the ultrafast propagation dynamics of femtosecond laser pulses in transparent materials, $CS_2$ and fused silica, respectively. Through consecutive snap-shot imaging of the laser pulses propagating in the media, ultrafast temporal changes in the spatial distribution of the optical pulse intensity were obtained. Based on the FTOP technique, a femtosecond time-resolved single-shot OKE measurement was demonstrated, which can be accomplished using a single-laser shot and has a femtosecond time resolution. The polarization dependence of the FTOP image intensity indicated that the FTOP images were mainly originated from transient birefringence effect induced by the pump laser pulse.

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## References

1. J. M. Dudley, G. Genty, and S. Coen, "Supercontinuum generation in photonic crystal fiber," *Rev. Mod. Phys.* **78**(4), 1135–1184 (2006).
2. V. P. Kandidov et al., "Self-transformation of a powerful femtosecond laser pulse into a white-light laser pulse in bulk optical media (or supercontinuum generation)," *Appl. Phys. B.* **77**(2–3), 149–165 (2003).
3. W. Liu et al., "Femtosecond laser pulse filamentation versus optical breakdown in H2O," *Appl. Phys. B.* **76**(3), 215–229 (2003).
4. F. Theberge et al., "Tunable ultrashort laser pulses generated through filamentation in gases," *Phys. Rev. Lett.* **97**(2), 023904 (2006).
5. J. Fuchs et al., "Dynamicss of subpicosecond relativistic laser pulse self-channeling in an underdense preformed plasma," *Phys. Rev. Lett.* **80**(8), 1658–1661 (1998).
6. M. P. Poudel and J. H. Chen, "Nonlinear optical effects during femtosecond photodisruption," *Opt. Eng.* **48**(11), 114302 (2009).
7. A. Pukhov and J. Meyer-ter-Vehn, "Relativistic magnetic self-channeling of light in near-critical plasma: three-dimensional particle-in-cell simulation," *Phys. Rev. Lett.* **76**(21), 3975–3978 (1996).
8. C. E. Clayton et al., "Plasma wave generation in a self-focused channel of a relativistically intense laser pulse," *Phys. Rev. Lett.* **81**(1), 100–103 (1998).
9. H. Kumagai et al., "Observation of the complex propagation of a femtosecond laser pulse in a dispersive transparent bulk material," *J. Opt. Soc. Am.* **20**(3), 597–602 (2003).
10. E. L. Falcao et al., "Ultrafast nonlinearity of antimony polyphosphate glasses," *Appl. Phys. Lett.* **83**(7), 1292–1294 (2003).
11. D. G. Kong et al., "Ultrafast third-order nonlinear optical properties of $ZnPc(OBu)_6(NCS)$/DMSO solution," *Opt. Lett.* **34**(16), 2471–2473 (2009).
12. J. F. He et al., "A signal to noise ratio measurement for single shot laser pulses by use of an optical Kerr gate," *Opt. Express* **19**(5), 4438–4443 (2011).
13. N. Furukawa et al., "Femtosecond real-time pump-probe imaging spectroscopy," *Appl. Phys. Lett.* **85**(20), 4645–4647 (2004).

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