Ultrafast imaging of photoinduced nonlinear response in transparent materials using femtosecond optical polarigraphy technique

Pengchao He
Jinhai Si
Lihe Yan
Xin Liu
Xiaofang Wang
Feng Chen
Xun Hou
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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.

Keywords: femtosecond techniques; ultrafast processes; nonlinear optics.

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.⁵ 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.⁸ 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 medium, 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...
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-μJ energy in our experi-
ments, 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
β-
barium borate (BBO) crystal. After passing a bandpass filter
centered at 400 nm, the probe beam is collimated and intro-
duced 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 sam-
ple, a polarizer (P1) is set to 45 deg with respect to the hori-
zontal 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
4
f
system is located on the imaging plane
of the filaments. The spatial resolution of the imaging system
is 3.4 μm/pixel.

3 Results and Discussion
First, we recorded the FTOP images of the pump pulse propa-
gating in CS₂ 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 CS₂ and fused silica, respectively. The pump power was 10 mW and propa-
gated from left to right, and time proceeds from top to bot-
tom. To increase the signal to noise ratio, the exposure time of the CCD camera for each image was set at 1/2 s, correspond-
ing 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 correspond-
ence 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 femto-
second 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 CS₂, respec-
tively. From the figure, we can see that the FTOP image in
CS₂ 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 CS₂ originated from the orientation of the
molecules has a long response time, the residual birefrin-
gence 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 CS₂.

The squares and circles in Fig. 3(c) show the normalized
intensity distribution of FTOP images along the axial direc-
tion recorded in CS₂ and fused silica, respectively. The hori-
zontal 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 CS₂, the nonlinear response showed an
obvious slow relaxation process. Using an exponential func-
tion, we fitted the decay processes of image intensity as

![Fig. 1 Experimental setup for optical polarigraphy. BS: beam splitter,
M: mirror, L: lens, P: polarizer, and HWP: half-wave plate, F: band-
pass filter.](image)

![Fig. 2 Images of a 10-mW pump pulse propagation in (a)–(d) CS₂ and (e)–(f) fused silica at different
time.](image)
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 pulse irradiation.$^{12}$ 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 $yz$ plane, it will induce a refractive index change on $x$ and $z$ axes, respectively. 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:

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

(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$^{13}$

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

(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, CS₂ 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

Pengchao He received his BE degree in electronic science and technology from Xi’an Jiaotong University, China, in 2011. He is currently pursuing his MS degree with a focus on ultrafast time-resolved pump-probe technique in materials.

Biographies of the other authors are not available.