Multi-Frame Observation of a Single Femtosecond Laser Pulse Propagation Using an Echelon and Optical Polarigraphy Technique

Lihe Yan, Xiaofang Wang, Jinhai Si, Pengchao He, Feng Chen, Jianhua Zou, and Xun Hou

***Abstract*— We developed a multiframe observation method of femtosecond laser pulse propagating in transparent medium based on optical polarigraphy technique. An echelon was introduced into the probe light and divided it into multipulses both in time and space, allowing a multiframe detection of the intense laser pulse propagating in materials. Using this method, we realized a multiframe observation of a single femtosecond laser pulse propagating in CS2 with ultrafast self-modulation such as filamentation. This imaging method has been demonstrated to be of a femtosecond time resolution and a frame rate of ∼THz.**

***Index Terms*— Optical imaging, ultrafast optics, nonlinear optics, Kerr effect.**

I. INTRODUCTION

IN THE past few decades, many methods have been developed to observe the propagation behavior of intense laser pulses in transparent media [1]–[3]. Femtosecond time-resolved optical polarigraphy (FTOP) technique, which uses the instantaneous birefringence induced by the strong electrical field of the pulse, can be used to measure the ultrafast temporal changes in the two-dimensional spatial distribution of the optical pulse intensity [4]–[6]. If a series of instantaneous intensity distributions at several successive temporal points are observed, however, the propagation profile will be quietly different from shot to shot due to the laser pulse fluctuation. To fulfill the single-shot detection of the pulse propagation dynamics, M. Fujimoto et al developed a successive four-frame instantaneous observation method of an intense pulse propagating in air [7]. This method used a quadruple-pulse generator in the optical path generating a quadruple-probe pulse, which captures successive four-frame instantaneous FTOP images of a single laser pulse. However, the quadruple-pulse generator which combines four different probe beams with different optical paths, will largely increase the complexity of the experiments.

Some other multi-frame measurements of the pulse propagation dynamics have been developed in the following years. M. Centuriona et al. used a segmented mirror arrays in the probe light path to create multiple holographic copies of the pulse propagating in water. By adjusting the position of the mirrors, successive multi-frame holograms were obtained [8]. Using a much simpler stair-step echelon, one can separate the probe light into several distinct beams, each with different temporal delay. Hence, pump-probe measurements can be accomplished using a single laser shot, or even a single ultra-short laser pulse, and with a fine time-resolution. In the previous reports, single-shot measurements using echelon have been explored in many pump-probe spectroscopy experiments [9]–[14]. For example, A. Nelson etc. observed the organic crystalline reaction dynamics in a single laser shot by implementing a two-dimensional spatial delay gradient across the profile of a femtosecond probe pulse using echelons [12].

In this letter, we developed a multi-frame observation method of femtosecond laser pulse propagating in transparent medium based on echelon and FTOP technique. An echelon was introduced into the probe light path and separated it into multi-pulse, allowing a multi-frame detection of the intense laser pulse propagating in materials. Using this method, we realized a four-frame observation of a single femtosecond laser pulse propagating in CS_2. Some ultrafast self-modulation effects such as filamentation and multi-filamentation were observed using a single laser shot.

II. EXPERIMENTS

Figure 1 illustrates the experimental setup for single-shot multi-frame FTOP imaging scheme based on echelon. A Ti: sapphire amplifier system 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 beam by a beam splitter. After passing through a delay line, the polarization of the pump beam is changed to vertical by 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 5 μJ energy in our experiments, the nonlinear focus is located at about 1 mm inside the input window of the cuvette.

Manuscript received May 21, 2013; revised July 29, 2013; accepted August 10, 2013. Date of publication August 16, 2013; date of current version September 11, 2013. This work was supported in part by the National Science Foundation of China under the Grant 61235003, in part by the National Basic Research Program of China (973 Program) under Grant 2012CB921804, and in part by the National Science Foundation of China under Grants 11304242 and 61205129. *(Corresponding author: J. Si.)*

L. Yan, X. Wang, J. Si, P. He, F. Chen, and X. Hou are with the Key Laboratory for Physical Electronics and Devices of the Ministry of Education and Shaanxi Key Laboratory of Information Photonic Technique, School of Electronics and Information Engineering, Xi'an Jiaotong University, Xi'an 710049, China (e-mail: jinhaisi@mail.xjtu.edu.cn).

J. Zou is with the School of Electronics and Information Engineering, Xi'an Jiaotong University, Xi'an 710049, China.

Color versions of one or more of the figures in this letter are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/LPT.2013.2278883

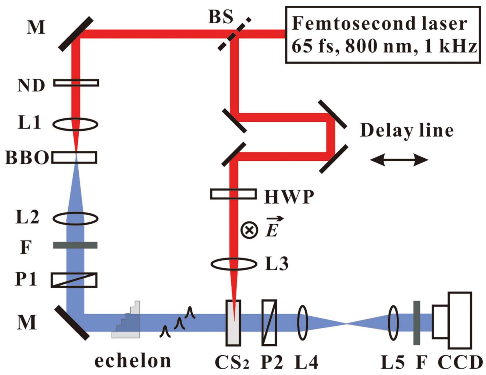

Fig. 1. Experimental setup of multi-frame FTOP imaging technique using an echelon. BS: beam splitter, M: mirror, ND: neutral density filter, L: lens, P: polarizer, HWP: half-wave plate, F: 400 nm bandpass filter.

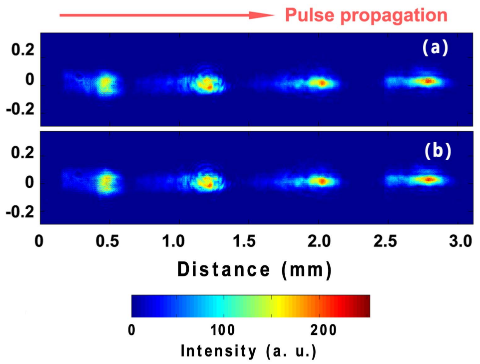

Fig. 2. Four-frame observation of a 5 μJ pulse propagating in CS_2 while it's being focused by a lens with a focal length of 100 mm. The pulse propagated from left to right. (a) and (b) Correspond to different laser shots under the same conditions.

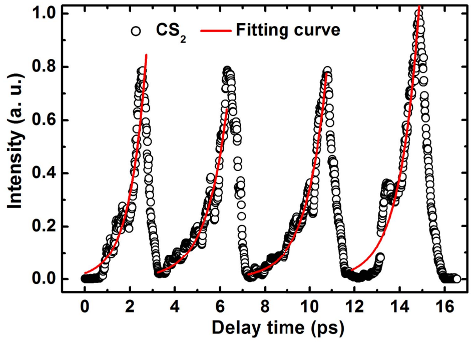

Fig. 3. Exposure intensity distribution of the four-frame FTOP image as a function of the propagation time of the pump pulse. The solid lines show the fitted curve of the decay process of CS_2.

To avoid the background scattering of the pump light, the probe beam is frequency-doubled after passing through a 1 mm thick BBO crystal. Then the probe light centered at 400 nm is collimated by a con-focal lens and passes through a stair-step echelon, which separates the probe light in space and time. The modulated probe light is introduced into the CS_2 cell perpendicularly to the direction of the pump path, with the light spot covering the area of the focal point of the pump beam. In front of the sample, a polarizer (P1) is set to 45 degree with respect to the horizontal plane of the optical stage and allows parts of the probe light to pass. When the probe pulse passes through the interaction region, it will experience the birefringence induced by the pump light and the linear polarization of the probe light will change to elliptical polarization. After passing through the crossed analyzer (P2) placed behind the sample, only the components perpendicular to the polarizer (P1) can be extracted. To record the polarigraphy image, a high-spatial-resolution charge coupled device (CCD) camera is located on the imaging plane of the pump light path.

The echelon with 4 steps is mechanically fabricated on a commercially available optical glass plate. The step width is 2 mm, producing a time delay of about 4.1 ps for 400 nm probe pulse ($n = 1.615$). As each step produces one frame of the FTOP image, the introduced delay time by each step must agree with the propagation time of the 800 nm pump pulse in CS_2 ($n = 1.62$). Hence, the height of each step of the echelon is designed to be 0.75 mm, with a width of 2 mm producing a delay time of about 4.1 ps.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the recorded four-frame instantaneous FTOP image of a 5 μJ pump pulse propagation in CS_2. The pulse propagated from the left to right. The exposure time of the CCD camera was set at 1 ms and a single laser pulse was used to produce the image. The spatial resolution of the image is estimated to be 2.2 μm/pixel. The image has four profiles each corresponding to one frame of the temporal intensity distributions of the laser pulse propagating in CS_2 at different time. The interval between the peaks of adjacent profiles was fixed at 0.75 mm, which was decided by the structure of the echelon. As each stair of the echelon had different thickness and transmittance, and the probe pulse intensity distribution was not strictly uniform, each frame of the FTOP image was normalized by comparing with the intensity distribution of the incident probe pulse after passing through the echelon. From the figure we can see that, the lateral size of the pump light spot changed slightly inside the sample even after the lens focus locating at 1 mm inside the sample. Because of the balance between Kerr self-focusing and plasma defocusing effect induced by the nonlinear ionization, a filament was produced inside CS_2 [15]. In addition, we recorded FTOP images for different laser shots under the same condition, as shown by Fig. 2(b). Due to the little fluctuation of the laser pulses, however, no obvious difference was observed in our experiments.

We acquired the exposure intensity distribution along the propagating axis of the FTOP profiles as given in Fig. 2(a). The circles in Fig. 3 show the acquired intensity distributions as a function of the propagating time of pump pulse. In the FTOP measurements, the intensity of recorded images is approximately proportional to the square of the pump light intensity, and the horizontal length of the image corresponds to the response time of the optical Kerr effect [16]. Using an exponential function, we fitted the decay processes of image intensity for each frame and an average exponential decay time of 0.87 ps was obtained, which was in the same

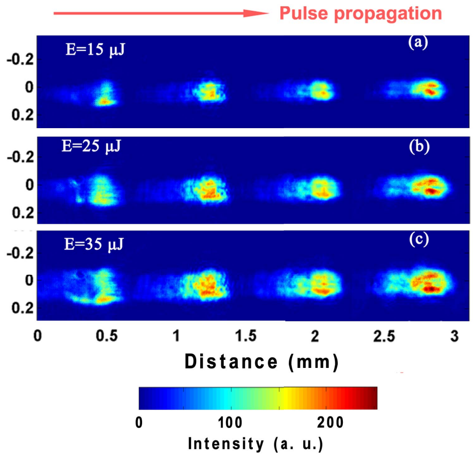

Fig. 4. Four-frame observation of a single pulse propagating in CS_2, with pulse energy of (a) 15 μJ, (b) 25 μJ, and (c) 35 μJ. The pulse propagated from left to right.

order with that in the previous reports [17]. The solid lines in Fig. 3 show the fitted relaxation process of the nonlinear response of CS_2, which was mainly attributed to the molecule reorientation. The frame interval was estimated to be about 4.05 ps, corresponding to a frame rate of about 0.25 THz. It should be noted that, the slow Kerr response of CS_2 limited the temporal resolution as well as the frame rate of the imaging. When this method is used to observe the propagation dynamics of an ultra-short pulse in materials with ultrafast response, such as air and glasses, the temporal resolution of the imaging will be mainly limited only by the pulse duration, and the frame rate of the imaging might be even higher [7], [16].

In addition, we observed the propagation dynamics of a single laser pulse in CS_2 with different pulse energy using the multi-frame FTOP method. Figures 4(a)–(c) indicate the recorded FTOP images of a pulse with energy of 15 μJ, 25 μJ, and 35 μJ, respectively. Similarly to Fig. 3, each panel reveals single pulse propagation, and the interval between the two adjacent profiles of the image was fixed at 0.75 mm. In Fig. 4, we can see the details of the pulse profile changing as it propagates. With increasing the pulse energy, the fine structures of the propagation profiles, such as lateral size and the numbers of filaments changed. Two filaments were formed when 15 μJ and 25 μJ pulse propagated in the medium, while three or more filaments with more complicated structures were formed in the 35 μJ pulse propagation path.

IV. CONCLUSION

In conclusion, we developed a multi-frame observation method of femtosecond laser pulse propagating in transparent medium based on FTOP technique. An echelon was introduced into the probe light and divided it into multi-pulses both in time and space, allowing a four-frame detection of the intense laser pulse propagating in materials. Using this method, we have succeeded in a successive four-frame observation of a single femtosecond laser pulse propagating in CS_2. This instantaneous imaging method has been demonstrated to be of a femtosecond time resolution and a frame rate of about 0.25 THz or even higher.

REFERENCES

[1] C. E. Clayton, *et al.*, "Plasma wave generation in a self-focused channel of a relativistically intense laser pulse," *Phys. Rev. Lett.*, vol. 81, no. 1, pp. 100–103, 1998.

[2] S. Minardi, *et al.*, "Time-resolved refractive index and absorption mapping of light-plasma filaments in water," *Opt. Lett.*, vol. 33, no. 1, pp. 86–88, 2008.

[3] T. Balciunas, A. Melninkaitis, G. Tamosauskas, and V. Sirutkaitis, "Time-resolved off-axis digital holography for characterization of ultrafast phenomena in water," *Opt. Lett.*, vol. 33, no. 1, pp. 58–60, 2008.

[4] M. Fujimoto, S. Aoshima, M. Hosoda, and Y. Tsuchiya, "Femtosecond time-resolved optical polarigraphy: Imaging of the propagation dynamics of intense light in a medium," *Opt. Lett.*, vol. 24, no. 12, pp. 850–852, 1999.

[5] M. Fujimoto, S. Aoshima, and Y. Tsuchiya, "Ultrafast imaging to measure instantaneous intensity distributions of femtosecond optical pulses propagating in a medium," *Meas. Sci. Technol.*, vol. 13, no. 11, pp. 1698–1709, 2002.

[6] I. Blonskyi, M. Brodyn, V. Kadan, O. Shpotyuk, I. Dmitruk, and I. Pavlov, "Spatiotemporal dynamics of femtosecond filament induced plasma channel in fused silica," *Appl. Phys. B*, vol. 97, no. 4, pp. 829–834, 2009.

[7] M. Fujimoto, S. Aoshima, and Y. Tsuchiya, "Multiframe observation of an intense femtosecond optical pulse propagating in air," *Opt. Lett.*, vol. 27, no. 5, pp. 309–311, 2002.

[8] M. Centurion, Y. Pu, and D. Psaltis, "Holographic capture of femtosecond pulse propagation," *J. Appl. Phys.*, vol. 100, no. 6, pp. 063104-1–063104-9, 2006.

[9] M. R. Topp, P. M. Rebtzepi, and R. P. Jones, "Time-resolved absorption spectroscopy in 10–12 sec range," *J. Appl. Phys.*, vol. 42, no. 9, p. 3415, 1971.

[10] G. P. Wakeham, D. D. Chung, and K. A. Nelson, "Femtosecond time-resolved spectroscopy of energetic materials," *Thermochim. Acta*, vol. 384, nos. 1–2, pp. 7–21, 2002.

[11] I. Katayama, H. Sakaibara, and J. Takeda, "Real-time time-frequency imaging of ultrashort laser pulses using an echelon mirror," *Jpn. J. Appl. Phys.*, vol. 50, pp. 102701-1–102701-5, Oct. 2011.

[12] P. R. Poulin and K. A. Nelson, "Irreversible organic crystalline chemistry monitored in real time," *Science*, vol. 313, no. 5794, pp. 1756–1760, 2006.

[13] 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*, vol. 19, no. 5, pp. 4438–4443, 2011.

[14] K. Y. Kim, B. Yellampalle, A. J. Taylor, G. Rodriguez, and J. H. Glownia, "Single-shot terahertz pulse characterization via two-dimensional electro-optic imaging with dual echelons," *Opt. Lett.*, vol. 32, no. 14, pp. 1968–1970, 2007.

[15] L. H. Yan, *et al.*, "Time-resolved single-shot imaging of femtosecond laser induced filaments using supercontinuum and optical polarigraphy," *Appl. Phys. Lett.*, vol. 100, no. 11, pp. 111107-1–111107-3, 2012.

[16] M. Fujimoto, S. Aoshima, M. Hosoda, and Y. Tsuchiya, "Analysis of instantaneous profiles of intense femtosecond optical pulses propagating in helium gas measured by using femtosecond time-resolved optical polarigraphy," *Phys. Rev. A*, vol. 64, no. 3, pp. 033813-1–033813-11, 2001.

[17] T. Hattori and T. Kobayashi, "Ultrafast optical Kerr dynamics studied with incoherent light," *J. Chem. Phys.*, vol. 94, no. 5, pp. 3332–3346, 1991.