SCIENTIFIC REPORTS

natureresearch

OPEN

Check for updates

Motion-picture recording of ultrafast behavior of polarized light incident at Brewster's angle

Mika Sasaki¹, Atsushi Matsunaka¹, Tomoyoshi Inoue¹, Kenzo Nishio² & Yasuhiro Awatsuji ³

Observing light propagation plays an important role in clarifying ultrafast phenomena occurring on femtosecond to picosecond time scales. In particular, observing the ultrafast behavior of polarized light is useful for various fields. We have developed a technique based on Polarization Light-in-Flight Holography, which can record light propagation as a motion picture that can provide information about the polarization direction. Here we demonstrate motion-picture recording of a phenomenon, which is characteristic of polarized light, by using the proposed technique. As a phenomenon, we adopted the behavior of a light pulse incident at Brewster's angle. We succeeded in recording the light reflection of specific polarized light by the proposed optical setup. The method of recording the motion-picture, reconstruction procedure, and the quantitative evaluation of the results are demonstrated.

Recently, applications of a pulsed laser operating in the picosecond or femtosecond regimes have been actively studied in various fields such as engineering¹⁻³, physics^{4,5}, and biology^{6,7}. Observing the propagation of a light pulse is important for developing various studies in these fields. There have been many studies on the techniques for observing light pulse propagation. For example, femto-photography⁸, compressed ultrafast photography⁹⁻¹¹, femtosecond time-resolved optical polarigraphy (FTOP)¹²⁻¹⁵, single-photon sensitive light-in-fight imaging^{16,17}, pump-probe method¹⁸⁻²¹, light-in-flight (LIF) holography²²⁻²⁸, and so on. Velten et al. showed motion pictures of a propagating light pulse around the object using a streak camera⁸. The streak camera is an ultrafast photon detection system that transforms the temporal profile of the light signal into the temporal profile by pulling photoelectrons with a sweep voltage. However, a synchronization with scanning light source and repetitive light pulses were required to capture a two-dimensional (2D) image. To overcome this limitation, compressed ultrafast photography⁹⁻¹¹ has been developed. The technique combines compressed sensing and a streak camera to capture motion pictures of light propagation without scanning procedure. Compressed ultrafast photography has been applied to various applications, including imaging of light pulse reflection and refraction, fluorescence lifetime imaging⁹, and real-time observing of a photonic Mach cone¹⁰. FTOP, based on the optical Kerr effect, can be used to observe the light pulse propagation¹²⁻¹⁴. However, FTOP requires repetitive light pulses, the profile of the light pulse propagation is different from shot to shot. Although a single-shot FTOP system¹⁵ was developed afterwards, the number of frames captured in one acquisition is limited by the trade-off between temporal resolution and the imaging field of view (FOV). Gariepy et al. used a SPADs (single-photon avalanche diodes) array of 32 pixels \times 32 pixels to observe light propagation¹⁶. SPAD pixel offers extreme sensitivity and picosecond temporal resolution. The propagating light pulse in the air and optical fibers were observed by the SPADs^{16,17}. The pump-probe technique is one of the most common methods to understand not only the ultrafast phenomena but also the light propagation. The light pulse propagation and terahertz Cherenkov wave generated by focusing femtosecond light pulse into lithium niobite (LiNbO₃) were observed by using the pump-probe technique^{20,21}. Compared with other techniques, LIF holography has the following advantages. First, it has a high temporal resolution. Second, it can record light propagation as a spatially and temporally continuous motion picture. Third, it can obtain three-dimensional images of a light pulse²⁷. Moreover, we have developed a technique that can simultaneously obtain polarization information²⁸, termed, Polarization Light-in-Flight Holography. Some ultrafast phenomena vary in their behavior depending on polarization state²⁹, and some occur in polarization-sensitive materials³⁰. Therefore, polarization properties are important for our increased understanding of such phenomena.

¹Department of Electronics, Graduate School of Science and Technology, Kyoto Institute of Technology, Matsugasaki Goshokaido-cho, Sakyo-ku, Kyoto, 606-8585, Japan. ²Advanced Technology Center, Kyoto Institute of Technology, Matsugasaki Goshokaido-cho, Sakyo-ku, Kyoto, 606-8585, Japan. ³Faculty of Electrical Engineering and Electronics, Kyoto Institute of Technology, Matsugasaki Goshokaido-cho, Sakyo-ku, Kyoto, 606-8585, Japan. [⊠]e-mail: awatsuji@ kit.ac.jp



Figure 1. Experimental results. The white arrows indicate the motion direction. We capture the different motion pictures simultaneously (Video S1). In addition, we separately show individual motion pictures as other video files (Video S2, Video S3, Video S4, and Video S5). (a) The pulse is in air. (b) The pulse has reached the glass and the reflected light pulse has appeared. (c–j) Propagation of the reflected light pulse can be observed. (e–j) Propagation of the refracted light pulse can be observed.

In this paper, we report motion-picture recording of a phenomenon that is characteristic of polarized light by using the Polarization Light-in-Flight Holography. As a subject, we chose a light pulse incident at Brewster's angle. The phenomenon of Brewster's angle is well-known, which was discovered about two hundred years ago^{31,32}. However, to the best of our knowledge, no one has observed the reflection of specific polarized light as a motion picture. Our experimental results are the first demonstration that the phenomenon is observed as motion pictures, instead of still pictures of the trajectory of the light rays. Moreover, we quantitatively evaluate the result obtained by the Polarization Light-in-Flight Holography for the first time.

Experimental Results

A short-pulsed laser is used as a light source in the recording procedure of the Polarization Light-in-Flight Holography. Different horizontal points on the holographic plate get the information about the subject (light pulse) generated at different times. On the other hand, different longitudinal areas on the holographic plate get information about polarization. In order to obtain the polarization information, we record the amounts of the four polarization components included in the subject. This is achieved by the interference between the subject (object light pulse) and one of the four reference light pulses that vary in polarization direction. Therefore, four motion pictures, characterizing each polarization component, are recorded. The amounts of the four polarization components correspond to the intensities of the four reconstructed images. The detailed method and experimental setup are described in the next sections.

Figure 1 shows the experimental results. They are the reconstructed images extracted from the recorded motion pictures at different time with different polarization components (see Video S1, Video S2, Video S3, Video S4, and Video S5). The numbers (0°-135°) on the right-hand side in Fig. 1 indicate each polarization component. The white lines show the air-glass interface. The recording time and the time interval between each image are 290 fs and 30 fs, respectively. The black lines shown in the reconstructed images indicate a background pattern. It was used for evaluating the magnification. It is observed that there is a little change between the 5th and 6th frames, as depicted in Fig. 1(e,f). However, there are remarkable changes between the 1st and 3rd frames and between the 8th and 10th frames, as depicted in Fig. 1(a-c,h-j).

Four reconstructed images, recorded at the same time, vary in intensity according to the polarization components. For example, regarding the incident light pulse, the reconstructed intensity of an image of the 45° linearly polarized component is the highest and that of the 135° linearly polarized component is the lowest, as depicted in Fig. 1. Moreover, regarding the reflected light pulse, the reconstructed image intensity of the 0° linearly polarized component is the highest, and that of the 90° linearly polarized component is too low to observe. From these results, it can be inferred that the polarization information of the light pulse incident at Brewster's angle can be obtained successfully.

Discussion

We evaluated the reconstructed image intensity obtained in the present study. Figure 2 shows the standardized pixel values of the images of the reflected light pulse. The numbers (0°-135°) on the right-hand side, in Fig. 2, indicate each polarization component. The yellow rectangles indicate the measuring areas, and their size is 300 pixels \times 80 pixels. The standardized pixel values of the 0°, 45°, 90°, and 135° linearly polarized components at their maximum are 0.93, 0.47, 0.047, and 0.46, respectively.

We quantitatively discussed the dependence of the reconstructed image intensity on polarization. Here, we defined the difference angle in polarization direction between the object light pulse and the reference light pulse as α . In conclusion, the relationship between the pixel values of the four reconstructed images and α is not linear. Because of the Malus's law³³, the pixel value of the reconstructed image depends on $\cos^2(\alpha)$. Assuming that the pixel value of the reconstructed image at $\alpha = 0^\circ$ is 1 and the pixel value of the reconstructed image at $\alpha = 45^\circ$ is



Figure 2. Standardized pixel values of the images of the reflected light pulse.

0.5. From the results of Fig. 2, it can be shown quantitatively that 0° polarized light selectively appears as reflected light, and this result matches the experimental condition.

Method

The Polarization Light-in-Flight Holography (the Polarization LIF Holography) is one of the ultrafast imaging techniques, from which it is possible to obtain motion pictures with polarization information of a propagating light pulse. It contains two-component techniques. One technique enables us to record light pulse propagation as a spatially and temporally continuous motion picture. The principle of this technique is the same as the usual LIF holography. The other enables us to visualize the polarization direction of a propagating light pulse. In order to identify polarization direction, we must obtain the intensity images of different polarized components. One of the powerful polarization imaging tools, the polarization camera^{34,35}, generally captures intensity images of four linear polarized components; therefore, we adopted the method of acquiring the four intensity images.

First, we explain the principle of the first component technique to record light pulse propagation as a spatially and temporally continuous motion picture. Figure 3(a) shows an optical setup of the Polarization LIF Holography. In the recording procedure, a short-pulsed laser is used as a light source. A light pulse emitted from the short-pulsed laser is divided into two light pulses by a beam splitter. One light pulse illuminates the object and it is called the object-illuminating light pulse. The other is called the reference light pulse. The object-illuminating light pulse is obliquely introduced to a diffuser plate with a certain incident angle. Then, the object-illuminating light pulse sweeps different horizontal points on the diffuser plate. The object-illuminating light pulse is scattered by the diffuser plate, then light pulses are generated from different points at different times. These light pulses are called the object light pulses. The reference light pulse is also obliquely introduced to a holographic plate with a certain incident angle, θ , and then sweeps different horizontal points on the holographic plate. The interference between the object light pulses and the reference light pulse occurs only when they arrive at a point on the



Figure 3. Schematic of the Polarization Light-in-Flight Holography. (**a**) Optical setup for the technique. M1-M4, mirrors; BS, beam splitter; QWP, quarter-wave plate; OB1 and OB2, objective lenses; CL1 and CL2, collimator lenses; and PFA, polarizing filter array. (**b**) The basic concept of the technique to visualize the polarization direction of a propagating light pulse.

holographic plate simultaneously. In other words, the different horizontal points on the holographic plate get the information about an object light pulse generated at different times. In the reconstruction procedure, a continuous wave (CW) laser is used for reconstructing the motion picture of the light pulse propagation. We choose a CW laser emits light whose wavelength is approximately the same as the center wavelength of the pulsed laser used to record the interference pattern or the holograms. The light emitted from the CW laser is collimated and illuminates the hologram at the angle, θ . By moving the gazed point on the hologram along the direction in which the reference light pulse swept the holographic plate, we can observe an optical image of a spatially and temporally continuous motion picture of the light pulse propagation.

Next, we explain the principle of the second component technique, i.e., visualization of the polarization direction of a propagating light pulse. The basic concept of the technique is one of the Fresnel-Arago Laws³⁶ stating that two orthogonal, coherent linearly polarized waves cannot interfere. A polarizing filter array (PFA) is introduced into the reference light pulse path. PFA is made by longitudinally arranging four pieces of linear polarizing film whose respective transmission axes are 0°, 45°, 90°, and 135°. The reference light pulse is changed to circularly polarized light pulse by a quarter-wave plate (QWP) before PFA. Thus, PFA gives longitudinal spatial distribution of four linear polarized light pulses to the reference light pulse. The holographic plate is divided longitudinally, and then we obtain four holograms that vary in the polarization direction of the reference light pulse. For example, we obtain four motion pictures, as shown in Fig. 3(b), when we record the propagation of a 0° linearly polarized light pulse. The object light pulses and the 0° linearly polarized reference light pulse interfere most



Figure 4. Experimental setup. (**a**) Optical setup for the experiment. M1-M5, mirrors; BS, beam splitter; QWP, quarter-wave plate; HWP, half-wave plate; OB1-OB3, objective lenses; CL1-CL3, collimator lenses; P, polarizer; and PFA, polarizing filter array. (**b**) Detail view of the object. (**c**) PFA, the polarizing filter array.

constructively. However, the object light pulses and the 90° linearly polarized reference light pulse do not interfere. The contrast of the interference fringes corresponds to the reconstructed image intensity. Thus, the reconstructed image intensity of the 0° linearly polarized component is the highest of the four images. On the other hand, the reconstructed image intensity of the 90° linearly polarized component is the lowest. The intensities of the reconstructed images of the 45° and 135° linearly polarized components are intermediate because the 45° and 135° linearly polarized reference light pulses include the 0° linearly polarized component. This is why we can obtain the polarization information of the object light pulse from the four reconstructed images.

Experimental Setup

Figure 4(a) shows the experimental setup. A mode-locked pulsed laser (HighQ-2 SHG, Spectra-Physics Inc.) was used for the light source. The pulse duration and the central wavelength were 178 fs and 522 nm, respectively. A Konica P-5600 was used as the recording medium of the interference (hologram). As shown in Fig. 4(b), the object-illuminating light pulse illuminated the object that was made with a diffuser plate, a glass block, and a transparent film. A USAF test target was printed on the film. The film was not set to evaluate the resolving power of this system but to notice the light pulse propagation easily. As shown in Fig. 4(c), PFA was made with four polarizing films, and the size of each was $2 \text{ mm} \times 40 \text{ mm}$. In order to observe the air-glass interface in detail, we placed a magnifying optical system into the object light pulse path. The object-illuminating light pulse and the reference light pulse were introduced from the opposite direction because images of the object light pulses were reversed up/down and left/right by the magnifying optical system. The object-illuminating light pulse was reflected diagonally upward by the mirror, M4 and it was incident on the upper-end surface of the glass block at Brewster's angle by the mirror, M5. The longitudinal section of the light pulse was thick because the object-illuminating light pulse was incident from the obliquely upward direction. When the longitudinal section of the light pulse is thick, it is not easy to observe light propagation. Therefore, a cylindrical lens was placed into the path of the object-illuminating light pulse in order to observe the shape of the light pulse clearly. By using a half-wave plate and a polarizer, the incident light pulse was adjusted to a 45° linearly polarized light pulse consisting of s-polarized and p-polarized components equally. The purpose of the adjustment is to record different behavior of differently polarized light. A Nd:YVO₄ laser emitting a CW light beam, whose wavelength is 532 nm, was used to reconstruct the motion picture. Thanks to the setup, we succeeded in observing the behavior of the light pulse incident at Brewster's angle as motion pictures, for the world's first time.

Received: 6 January 2020; Accepted: 19 April 2020; Published online: 06 May 2020

References

- 1. Bauer, F., Michalowski, A., Kiedrowski, T. & Nolte, S. Heat accumulation in ultra-short pulsed scanning laser ablation of metals. *Opt. Express* 23, 1035–1043 (2015).
- 2. Harzic, R. L. et al. Sub-100 nm nanostructuring of silicon by ultrashort laser pulses. Opt. Express 13, 6651-6656 (2005).
- 3. Itoh, K., Watanabe, W. & Ozeki, Y. Nonlinear ultrafast focal-point optics for microscopic imaging, manipulation, and machining. *Proc. IEEE* 97, 1011–1030 (2009).
- 4. Choi, Y. *et al.* Measurement of the time-resolved reflection matrix for enhancing light energy delivery into a scattering medium. *Phys. Rev. Lett.* **111**, 243901 (2013).
- Yasui, T. et al. Adaptive sampling dual terahertz comb spectroscopy using dual free-running femtosecond lasers. Sci. Rep 5, 10786 (2015).
- 6. Watanabe, W. et al. Femtosecond laser disruption of subcellular organelles in a living cell. Opt. Express 12, 4203–4213 (2004).
- König, K., Schenke-Layland, K., Riemann, I. & Stock, U. A. Multiphoton autofluorescence imaging of intratissue elastic fibers. Biomaterials 26, 495–500 (2005).
- Velten, A. *et al.* Femto-photography: capturing and visualizing the propagation of light. *ACM Trans. Graph* 32, 1 (2013).
 Gao, L., Liang, J., Li, C. & Wang, L. V. Single-shot compressed ultrafast photography at one hundred billion frames per second. *Nature* 516, 74–77 (2014).
- Liang, J. et al. Single-shot real-time video recording of a photonic Mach cone induced by a scattered light pulse. Sci. Adv 3, e1601814 (2017).
- 11. Liang, J., Zhu, L. & Wang, L. V. Single-shot real-time femtosecond imaging of temporal focusing. Light Sci. Appl 7, 42 (2018).
- 12. Hosoda, M., Aoshima, S., Fujimoto, M. & Tsuchiya, Y. Femtosecond snapshot imaging of propagating light itself. Appl. Opt. 41, 2308–2317 (2002).
- Fujimoto, M., Aoshima, S., Hosoda, M. & Tsuchiya, Y. Femtosecond time-resolved optical polarigraphy: imaging of the propagation dynamics of intense light in a medium. Opt. Lett. 24, 850–852 (1999).
- Kumagai, H. et al. Observation of the complex propagation of a femtosecond laser pulse in a dispersive transparent bulk material. J. Opt. Soc. Am. B 20, 597–602 (2003).
- 15. Wang, X. et al. High-frame-rate observation of single femtosecond laser pulse propagation in fused silica using an echelon and optical polarigraphy technique. Appl. Opt. 53, 8395 (2014).
- 16. Gariepy, G. et al. Single-photon sensitive light-in-fight imaging. Nat. Commun. 6, 1-7 (2015).
- 17. Warburton, R. et al. Observation of laser pulse propagation in optical fibers with a SPAD camera. Sci. Rep 7, 43302 (2017).
- Cheng, K. et al. Ultrafast dynamics of single-pulse femtosecond laser-induced periodic ripples on the surface of a gold film. Phys. Rev. B 98, 184106 (2018).
- Mouskeftaras, A., Guizard, S., Fedorov, N. & Klimentov, S. Mechanisms of femtosecond laser ablation of dielectrics revealed by double pump-probe experiment. *Appl. Phys. A* 110, 709–715 (2013).
- 20. Word, R. C., Fitzgerald, J. P. S. & Könenkamp, R. Light propagation and interaction observed with electrons. Ultramicroscopy 160, 84–89 (2016).
- Wang, Z., Su, F. & Hegmann, F. A. Ultrafast imaging of terahertz Cherenkov waves and transition-like radiation in LiNbO₃. Opt. Express 23, 8073 (2015).
- 22. Abramson, N. Light-in-flight recording by holography. Opt. Lett. 3, 121-123 (1978).
- 23. Abramson, N. Light-in-flight recording: high-speed holographic motion pictures of ultrafast phenomena. *Appl. Opt.* 22, 215–232 (1983).
- 24. Abramson, N. Optical fiber tested using light-in-flight recording by holography. Appl. Opt. 26, 4657 (1987).
- 25. Kubota, T. & Awatsuji, Y. Observation of light propagation by holography with a picosecond pulsed laser. *Opt. Lett.* **27**, 815–817 (2002).
- Yamagiwa, M., Komatsu, A., Awatsuji, Y. & Kubota, T. Observation of propagating femtosecond light pulse train generated by an integrated array illuminator as a spatially and temporally continuous motion picture. Opt. Express 13, 3296–3302 (2005).
- Kubota, T., Komai, K., Yamagiwa, M. & Awatsuji, Y. Moving picture recording and observation of three-dimensional image of femtosecond light pulse propagation. Opt. Express 15, 14348–14354 (2007).
- 28. Inoue, T. et al. Spatiotemporal observations of light propagation in multiple polarization states. Opt. Lett. 44, 2069–2072 (2019).
- 29. Yasumaru, N., Miyazaki, K. & Kiuchi, J. Femtosecond-laser-induced nanostructure formed on hard thin films of TiN and DLC. Appl Phys A 76, 983–985 (2003).
- Shin, S., Lee, K., Yaqoob, Z., So, P. T. C. & Park, Y. Reference-free polarization-sensitive quantitative phase imaging using singlepoint optical phase conjugation. Opt. Express 26, 26858–26865 (2018).
- 31. Brewster, B. On the laws which regulate the polarization of light by reflection from transparent bodies. *Proc. Royal Soc. Lond.* 2, 14–16 (1815).
- 32. Brewster, B. IX. On the laws of the polarization of light by refraction. Phil. Trans. R. Soc 120, 133-144 (1830).
- 33. Gladden, S. C. An experiment on Malus' law for the elementary laboratory. Am. J. Phys 18, 395 (1950).
- 34. Gruev, V., Perkins, R. & York, T. CCD polarization imaging sensor with aluminum nanowire optical filters. Opt. Express 18, 19087–19094 (2010).
- Onuma, T. & Otani, Y. A development of two-dimensional birefringence distribution measurement system with a sampling rate of 1.3 MHz. Opt. Commun. 315, 69–73 (2014).
- 36. Arago, F & Fresnel, A. Memoir on the action of rays of polarized light upon each other in *The Wave Theory of* Light (ed. Crew, H.) 145–157 (American, 1900).

Acknowledgements

Japan Society for the Promotion of Science (JSPS) KAKENHI Grant-in-Aid for Scientific Research (A) (17H01062).

Author contributions

M.S. and A.M. fabricated the system and performed the experiments. All authors discussed the results and contributed to the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41598-020-64714-w.

Correspondence and requests for materials should be addressed to Y.A.

Reprints and permissions information is available at www.nature.com/reprints.

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2020