Single-shot 25-frame burst imaging of ultrafast phase transition of Ge\textsubscript{2}Sb\textsubscript{2}Te\textsubscript{5} with a sub-picosecond resolution

Takakazu Suzuki\textsuperscript{1}, Ryohei Hida\textsuperscript{1}, Yuki Yamaguchi\textsuperscript{1}, Keiichi Nakagawa\textsuperscript{2,3,4}, Toshiharu Saiki\textsuperscript{1}, and Fumihiko Kannari\textsuperscript{1*}

\textsuperscript{1}Department of Electronics and Electrical Engineering, Keio University, Yokohama 223-8522, Japan
\textsuperscript{2}Department of Bioengineering, The University of Tokyo, Bunkyo, Tokyo 113-8656, Japan
\textsuperscript{3}Department of Precision Engineering, The University of Tokyo, Bunkyo, Tokyo 113-8656, Japan
\textsuperscript{4}Medical Device Development and Regulation Research Center, The University of Tokyo, Bunkyo, Tokyo 113-8656, Japan

E-mail: kannari@elec.keio.ac.jp

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We captured ultrafast two-dimensional (2D)-burst images of the crystalline-to-amorphous phase transition of Ge\textsubscript{2}Sb\textsubscript{2}Te\textsubscript{5}. These transitions were induced by a femtosecond laser pulse, and the images, with a sub-picosecond temporal resolution, were acquired on a single-shot basis through the change in local optical transmittance. We employed a 2D-burst imaging method of sequentially timed all-optical mapping photography utilizing spectral filtering (SF-STAMP). The SF-STAMP system consists of a 25-beam-generating diffractive optical element, a band-pass filter, and two Fourier transform lenses. We used a frequency-chirped broadband pulse and achieved 25-frame burst imaging with an interval of 133 fs in a single-shot time window of 3.2 ps.© 2017 The Japan Society of Applied Physics

For capturing and elucidating ultrafast phenomena with a temporal resolution less than one nanosecond, the following methods have been developed: a pump-probe method using ultrafast laser pulses, a streak camera (whose highest temporal resolution is less than 1 ps), and an ultrahigh speed camera (up to $2 \times 10^8$ fps).\textsuperscript{1-5} However, the pump–probe laser pulse method, where the temporal resolution is determined by the pulse width of a probe laser, needs repetitive measurements. Therefore, the motion of non-repetitive or random events cannot be reconstructed. Although a streak camera can capture single events on a single-shot basis with sub-picosecond temporal resolution, the acquired image is only in one dimension. In recent years, for consecutive or single-shot imaging in the ultrashort time regions, two-dimensional (2D) ultrafast imaging methods have been developed, including optical time-stretch imaging,\textsuperscript{6,7} compressed ultrafast photography (CUP),\textsuperscript{8-10} sequentially timed all-optical mapping photography (STAMP),\textsuperscript{11} and others.\textsuperscript{12-14} STAMP is currently the world’s fastest 2D-burst imaging technique and can directly capture 2D-burst images of ultrafast phenomena with sub-picosecond temporal resolution. In STAMP, the snapshot of spectral images measured by a linearly frequency-chirped laser pulse corresponds to the temporal snapshot. However, the number of unique periscopes used in the original STAMP scheme limited the total number of frames (the original STAMP setup achieved six frames\textsuperscript{11}). To increase the number of STAMP frames by a factor of four, we introduced a scheme utilizing a “spatially and temporally resolved intensity and phase evaluation device: full information from a single hologram (STRIPED FISH)”\textsuperscript{15-17} method to STAMP and successfully demonstrated five-frame burst imaging by STAMP utilizing spectral filtering (SF-STAMP).\textsuperscript{18,19}

In the present study, we captured ultrafast 2D-burst images of the crystalline-to-amorphous phase transition of Ge\textsubscript{2}Sb\textsubscript{2}Te\textsubscript{5} (GST) with sub-picosecond temporal resolution on a single-shot basis for the first time, by improving the SF-STAMP setup to acquire 25 frames in a single-shot measurement. GST can undergo repeated phase changes between its crystalline and amorphous phases. These optical properties have been used in rewritable optical recording media such as DVDs.\textsuperscript{20} In optical recording systems, reversible phase changes between crystalline and amorphous phases are induced by irradiating the material with focused nanosecond laser pulses or continuous-wave laser output, which causes transient temperature ramping. On the other hand, in recent years, ultrafast crystalline-to-amorphous phase transitions induced by femtosecond laser pulses by a non-thermal process, which occurs in the sub-picosecond time regime, have often been reported.\textsuperscript{21-26} Time-resolved methods that rely on X-ray and electron diffraction and single-shot spectroscopy have been employed to perform these measurements. However, no results have been reported with 2D-image measurements for this ultrafast phase transition in a single-shot measurement scheme.

Figure 1 shows the schematic setup of an SF-STAMP system\textsuperscript{27} whose 4\textsuperscript{th} imaging configuration consists of a diffractive optical element (DOE) and a band-pass filter (BPF). The probe beam is optically Fourier transformed by the first lens, and then the beam is duplicated by the DOE to form plural array beams. The tilted BPF spectrally resolves the diffracted array beams depending on their incident beam angles. The spectrally filtered beams are optically inverse Fourier transformed by the second lens, and 2D-multispectral images are simultaneously generated on the image plane. When using a linearly frequency-chirped pulse, the spectral image corresponds to a snapshot at an ultrashort observation time slot in the chirped probe pulse. Since the number of frames in SF-STAMP is determined by the beam-generating DOE, increasing this number is easy. Therefore, the SF-STAMP system is simpler and more flexible than the original STAMP.\textsuperscript{11}

To perform our experiment, we used a specially designed 25-beam-generating DOE (HOLOEYE) that generates $5 \times 5$ beams.
In our setup, these parameters were pulse and location of 25 BPF spectrally resolve the image, and (c) spectral properties of a broadband probe pulse and location of 25 BPF filter bands in the spectrum.

Fig. 2. (a) Design of the 25-beam-generating DOE (800 nm wavelength), (b) arrangement of a 25-beam-generating DOE and a BPF to spatially and spectrally resolve the image, and (c) spectral properties of a broadband probe pulse and location of 25 BPF filter bands in the spectrum.

Fig. 3. Twenty-five multispectral images captured by the SF-STAMP system.

Fig. 4. Experimental setup of single-shot system for measuring the ultrafast crystalline-to-amorphous phase transition in GST.

\[
\tau = \sqrt{\frac{2\lambda_0^2 \ln 2}{\pi c (\Delta \lambda_{BPF})^2} + (D \cdot z \cdot \Delta \lambda_{BPF})^2},
\]

\[
\Delta t = D \cdot z \cdot \Delta \lambda_{DOE}.
\]

Here, \( \Delta \lambda_{BPF} \) is the spectral bandwidth (FWHM) of a BPF, \( \Delta \lambda_{DOE} \) is the difference between neighboring spectra, \( \lambda_0 \) is the center wavelength of the laser pulse from the optical source, and \( c \) is the speed of light in a vacuum. Equation (3) indicates that the temporal resolution of each frame shot increases as the spectral width of the BPF narrows with the second term; however, if the spectral width is too narrow, the exposure time increases because of the relation of the Fourier transform with the first term. Therefore, an optimum BPF spectral width exists that can maximize the temporal resolution. In our system, where \( \Delta \lambda_{BPF} = 2.2 \text{ nm} \), \( \Delta \lambda_{DOE} = 1.7 \text{ nm} \) (on average), and \( \lambda_0 = 805 \text{ nm} \), the averaged values of \( \tau \) and \( \Delta t \) were 465 and 133 fs, respectively.

The entire experimental setup is shown in Fig. 4. The light source was a mode-locked Ti:sapphire laser (Coherent Mira) with chirped pulse amplification (CPA). The average energy was 1 mJ at a repetition rate of 1 kHz with a pulse width of 50 fs (FWHM). The amplified laser pulse (800 nm) was split into a pump pulse, which initiates the phase transformation of the GST film, and a probe pulse, which was focused into the Ar-gas-filled hollow-core fiber (HCF; 126-µm core diameter). In the HCF, a broadband pulse (650–900 nm) was generated...
by self-phase modulation and four-wave mixing. To stretch the pulse width, the broadband pulse was passed through optical glass rods (N-SF11 and BK7, which have dispersion parameters at 800 nm of $-551.8$ ps/nm·km and $-131.4$ ps/nm·km, respectively). The entire material dispersion $D \cdot z$ is 0.082 ps/nm. To avoid crosstalk between the spectra of the scattered pump and probe pulses, we converted the pump pulse to its second harmonic (400 nm) with a $\beta$-barium borate (BBO) nonlinear crystal. Moreover, the polarizations of the pulses were also adjusted to be orthogonal to each other. The GST sample was composed of a 0.3-mm-thick glass substrate, a 10-nm-thick GST film layer, and a 10-nm-thick SiO$_2$ protective coat. The pump laser fluence that is required to cause a permanent amorphous phase change is $\sim 10$ mJ/cm$^2$; thus, the pump pulse was focused on the GST sample from the direction opposite to that of the probe pulse using an objective lens ($\times 20$, NA = 0.40, Olympus LMPLFLN20x). To observe the GST phase transition, we detected only the intensity change of the transmitted probe laser. We constructed a transmission microscopic optical system and relayed an enlarged image to the SF-STAMP system. The intensity of the probe laser pulse was set below the threshold at which no phase change caused by the probe itself can occur. The glass plate with the GST sample was positioned at a slight angle to avoid being affected by the stray light due to reflection from the GST sample layers. For a single-shot measurement, we employed an optical chopper and a shutter to select one pulse.

We achieved synchronization between the pump and chirped probe pulses in two steps. First, at the sample position, we placed a BBO crystal and generated the sum-frequency with the Fourier transform limited pump pulse (800 nm in this case) and the chirped probe pulse. The wavelength of the sum-frequency pulse was measured by varying the relative delay between the two pulses and calibrating the position of the delay stage. We confirmed that the measured chirp rate agreed well with that estimated from the material dispersion of the glass rods. Next, we positioned the GST sample and converted the pump pulse to a frequency-doubled pump pulse. The optical path length of the BBO nonlinear crystal was compensated by the delay stage.

Figure 5 shows the result of an ultrafast crystalline-to-amorphous phase transition of GST induced by a 400 nm femtosecond laser excitation with an SF-STAMP set up for a five-frame capture. In this setup, we added the total dispersion of 0.082 ps/nm on the probe pulse. In this five-frame measurement, the spectral bandwidth in a single shot was 20 nm (from 818 to 804 nm), and the observation time window was 1.1 ps. The specific time for each frame is determined by the center frequency of the BPF and the linear chirp rate of the probe laser pulse. The average frame intervals were 287 fs corresponding to frame rates of 1.23 Tfps.

The result of the 25-frame SF-STAMP measurement is shown in Fig. 6. The single-shot time window was 3.2 ps, corresponding to a 40 nm bandwidth, and the average frame interval was 133 fs, corresponding to frame rates of 7.52 Tfps. In the cases of both Figs. 5 and 6, the average exposure time for each frame was 465 fs. Each wavelength image occupied $400 \times 300$ pixels. All the images were trimmed, the intensity differences were taken from the image obtained before the pump irradiation, and their brightness was adjusted to compensate for the intensity variation among the probe spectra.
Since we can ignore the dependence of the optical transmission of GST on wavelength within the probe bandwidth of 40 nm, no further correction was made for each frame image.

As shown in Figs. 5 and 6, within the area irradiated by the pump laser pulse, the GST film showed higher transmittance of the probe laser beam, and a bright amorphous mark appeared at ~250 fs after the pump laser irradiation. The minimum transmittance change detectable with the electrically cooled CCD camera was ~3% in our setup. The gradual change in the probe laser transmission up to ~660 fs is clearly shown in Fig. 6, but it was not resolved in the five-frame shot in Fig. 5. These single-shot measurement results agree with the phase transition time scale reported in previous research.21–23) The interference fringes in the pictures were caused by the diffraction of the pump pulse at the edge of the objective lens. The amorphized marks did not change after 798 fs and still exhibited the interference fringe. In comparison with this amorphized area, the surrounding crystalline areas retained high reflectance. Therefore, we confirmed that the spatial intensity distribution of the pump laser pulse is clearly coincident with the phase change pattern. The phase change domain does not spatially spread to the surrounding area. In the theory, which has been widely accepted, that laser-induced nonthermal amorphization is initiated by Ge-atom displacements from octahedral to tetrahedral sites.22) Therefore, the amorphized domain is defined by the pump irradiation area of the pump pulse, and even the interference pattern is transferred to amorphization.

Our 2D-burst result is the first observation of such a fast phase transition induced by a femtosecond laser pulse on a single event basis. On the other hand, Kobayashi et al. observed amorphization of GST with accumulated laser irradiation at a pulse repetition rate of 500 Hz by using spectrally encoded single-shot measurements.25) Similar repetitive measurements with our SF-STAMP scheme will be obtained when a 2D-image sensor with higher frame rates (>500 fps) is available. However, for the present CCD image sensors with a larger diameter as our setup, the frame rate is still 0.7 fps or less.

In summary, we modified the initial SF-STAMP system, expanded the probe laser bandwidth from 20 to ~40 nm, and successfully increased the number of frames from 6 to 25. As a 25-frame demonstration, we captured ultrafast 2D-burst images of the crystalline-to-amorphous phase transition of GST with a frame interval of 133 fs in an entire time window of 3.2 ps. The ultrafast phase transition of GST in a time scale of ~650 fs was captured on a single event basis for the first time. Although the light utilization efficiency of a probe pulse is lower in the SF-STAMP method than that in the original STAMP because of the beam duplication and spectral filtering, it is advantageous that the selected spectrum band can be easily varied by adjusting the incident angles to the BPF.

Many interesting ultrafast phenomena exist (such as interactions with shock waves) that lie outside the time-range of conventional ultrafast cameras. To apply SF-STAMP for ultrafast 2D-burst imaging to these phenomena, the time window must be extended to the sub-nanosecond region. When a much broader probe pulse such as an octave span supercontinuum pulse is used, such a chirped long pulse can be prepared. Since a single tilted BPF is no longer applicable, we should prepare an individual BPF array to select discrete spectral images. On the other hand, when applying a unique optical setup that can generate delayed pulses with different colors, or dispersive optics such as free-space angular-chirp-enhanced delay (FACED),26) longer chirped pulses can be generated even for pulses with a narrower spectrum. Since the chromatic characteristics of an object cannot be considered with the SF-STAMP measurement, the latter approach with its inherently narrower bandwidth may prove suitable for future development.

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