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Development of ultrafast time-resolved dual-comb spectroscopy

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Ultrafast time-resolved dual-comb spectroscopy (TR-DCS) has been demonstrated, which enables direct observations of transient responses of complex optical spectra by combining dual-comb spectroscopy with the pump–probe method. TR-DCS achieves two-dimensional spectroscopy with a wide dynamic range for both the temporal and frequency axes. As a demonstration, we investigated the femtosecond relaxation dynamics of a photo-excited InGaAs saturable absorber in the near-infrared frequency region. The transient response of the interferogram was successfully obtained, and both the amplitude and phase spectra of the dynamic complex transmittance were independently deduced without using the Kramers-Kronig relations. A high phase resolution in the order of milliradian was achieved by suppressing the effect from the slow phase drift caused in the experimental system. Our proof-of-principle experiment promotes a pathway to coherent, highly accurate, and multi-dimensional pump–probe spectroscopy using the optical frequency comb technology. [http://dx.doi.org/10.1063/1.4976730]

Measurement methods using precisely controlled optical frequency combs have recently attracted much attention in the field of metrology.[1] In particular, a dual-comb source with a slightly different repetition rate is a powerful tool, producing, with high accuracy, an automatically sweeping pulse train pair. Essentially, the precise controllability of the dual-comb realizes pulse scanning that is unprecedentedly rapid, long range, low jitter, and highly coherent. Dual-comb spectroscopy (DCS) is one of the most successful applications of the dual-comb technique,[2–7] which utilizes its great characteristic as a scanner in an interferometric spectrometer, to enable a rapid and precise spectral detection in a wide frequency range. DCS has exhibited great potential in a variety of fields, such as high-precision spectroscopy for molecules[3,4] and nonlinear spectroscopy.[5–7] Meanwhile, asynchronous optical sampling (ASOPS) has been known as another application of the dual-comb technique,[8–10] which utilizes it as a scanner in the pump–probe method. ASOPS enables the rapid acquisition of transient responses with a considerably wide temporal dynamic range and has been applied in the detection of coherent phenomena[8,9] and terahertz time-domain spectroscopy,[10] among others.

The dual-comb configuration can be treated as a rapid scanning method, able to carry out new measurements with an unprecedentedly wide dynamic range. Therefore, an extension of the dual-comb for multi-dimensional measurements can naturally be reminded as one of the further applications. However, to our knowledge, almost all dual-comb experiments reported so far have been limited to one-dimensional measurements. Here, we propose a "time-resolved dual-comb spectroscopy (TR-DCS)" system as an extension for multi-dimensional measurements, which is a combination of conventional DCS and the pump–probe method, enabling femtosecond time-resolved measurements of the fine spectra of complex optical properties.

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Investigating the dynamic complex properties, including phase information, is essential to comprehend the complete picture of ultrafast optical responses in materials just after photo-excitation. Such information has been studied via time-resolved interference spectroscopy.[11–13] In conventional methods, however, the active mechanics, required for imposing a delay in the interferometer, may cause some difficulties. Examples include spatial errors caused by moving mirrors, slow data acquisition, the necessity of a complex phase stabilization system, and a limited time-scanning range. Conversely, since DCS does not inherently include any mechanics that imposes a delay in obtaining an interference signal, TR-DCS is expected to achieve improved time-resolved interference spectroscopy.

There are several previous reports from the viewpoint of dynamic phenomenon observations using optical frequency combs. For example, Fleisher *et al.* studied the microsecond dynamic chemical reaction using comb spectroscopy with a virtually imaged phased array.[14] As with other examples, such as observing picosecond phenomena with DCS, a short-time Fourier transform (STFT) was applied to the free induction decay signal from a molecular sample.[4] However, in the STFT scheme, the temporal resolution is restricted due to the Fourier limit. In order to access the faster time region with DCS, it is necessary to combine a pump–probe method as proposed in this study.

In this letter, we demonstrate a TR-DCS system, which enables direct observations of the ultrafast transient complex optical spectrum by combining conventional DCS with the pump–probe method. As a demonstration, the femtosecond relaxation dynamics of a photo-excited InGaAs saturable absorber is investigated, and we show that both the transient amplitude and phase spectra of the complex transmittance can be independently deduced.

Figure 1(a) shows the concept of the TR-DCS system proposed here, which corresponds to the combination of DCS with the pump–probe method. In conventional DCS, only the signal and local combs are involved. The signal comb stores information of the sample optical properties and the local comb retrieves it as an interferogram (IGM) in the time domain. Through Fourier analysis, both the real and imaginary parts of the complex optical properties can be directly and rapidly obtained in a wide frequency range with an ultra-precise resolution. In the TR-DCS system, the pump comb, synchronized to the signal comb, is additionally introduced. By performing DCS with scanning the pump delay time, τ, the ultrafast transient response of the interferogram can be obtained and further Fourier analysis can provide the complex optical property dynamics of the sample. In this way, TR-DCS achieves two-dimensional spectroscopy in time vs frequency with a wide dynamic range for both the temporal and frequency axes.

For demonstrating TR-DCS, we constructed an experimental system as shown in Fig. 1(b). Two lab-built Er-fiber oscillators, mode-locked via nonlinear polarization rotation, were used as light sources. Their center frequencies and the repetition rates, f_{rep}, were approximately 192 THz (1560 nm) and 56 MHz, respectively. For the tight phase-locking of f_{rep} and the offset frequency, f_{ceo}, of the dual-comb, we constructed a feedback frequency control system using a 1560-nm slave continuous-wave laser (Planex, Redfern Integrated Optics) and a microwave reference based on the global positioning system. Here, a sufficiently narrow relative linewidth between the comb modes, on the order of MHz, was achieved using an intracavity electronic optical modulator integrated only in the local comb; see Ref. 15 for details on our phase-locking system. The frequency difference, Δf_{rep}, was set to approximately 120.6 Hz.

As shown in Fig. 1(b), the output from one comb oscillator was enhanced by an Er-doped-fiber amplifier, producing pulses with about 60 fs duration, which limited the temporal resolution of the time-resolved experiment. The amplified beam was divided into pump and probe beams with a polarization beam splitter, and the power split ratio was adjusted with a half-wave plate in front of the splitter. The pump pulse excited the sample with approximately 30 mW of power and a 150 μm spot diameter, and a variable delay, τ, was induced by a mechanical stage. The probe pulse, which played the role of the signal comb in the DCS, was divided again into the sample and reference paths. The pulses propagating through the sample path stored the properties of the target material and were spatially overlapped with the pulses coming from the shorter reference path. The reference pulse was utilized as the phase standard in the phase compensation analysis explained later. Each transmitted signal comb pulse was spatially combined with another comb, the slightly asynchronized local comb with the duration of a few picoseconds, due to the chirp in the fiber. The IGM was observed using

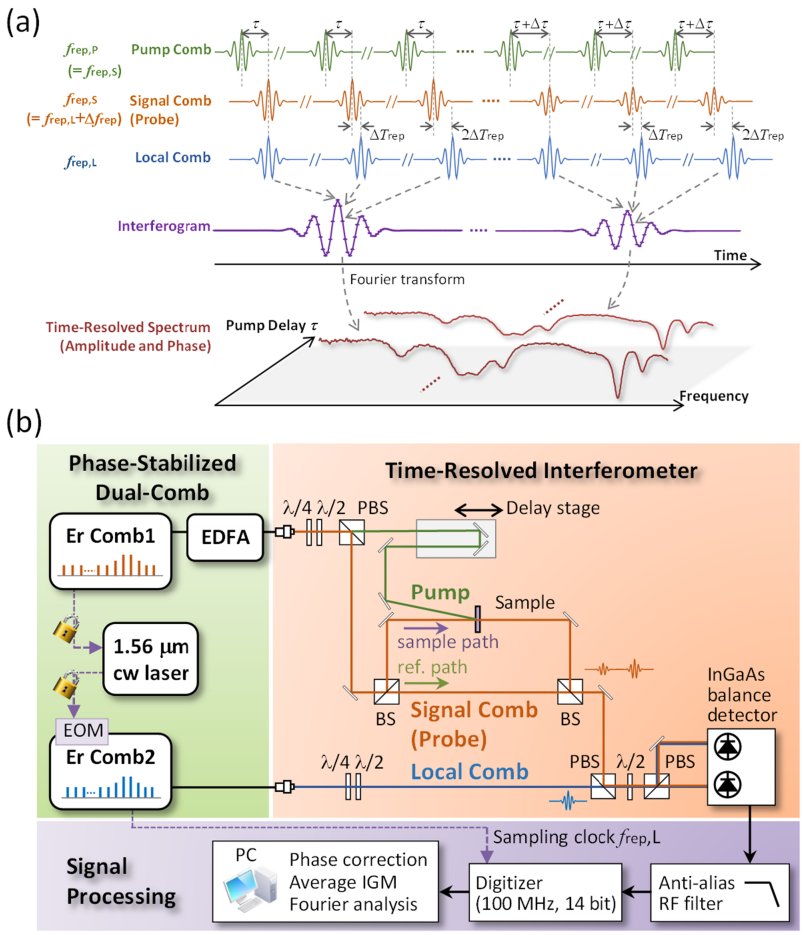

FIG. 1. (a) Concept of the time-resolved dual-comb spectroscopy (TR-DCS) system. (b) Schematic of the developed experimental system for the TR-DCS system. EDFA—Er-doped fiber amplifier; EOM—electro-optic modulator; PBS—polarization beam splitter; IGM—interferogram.

a balance detection system, in which the orthogonally polarized signal and local pulses were 45° rotated at a half-wave plate, and the interference signal without the dc component was obtained using an InGaAs balance detector (80 MHz bandwidth). The data were recorded by a digitizer (100 MHz bandwidth, 14-bit resolution) synchronized with the local comb.

As a demonstration of the TR-DCS system, we investigated the ultrafast relaxation dynamics after the photo-excitation of a commercial transmission type saturable absorber composed of InGaAs multi-quantum-wells (SA-1550-46-2ps, BATOP GmbH).

Figure 2(a) shows the IGM, which was observed in the condition that the pump pulses were incident to the sample 2 ps later than the probe (signal comb) pulses ($\tau = -2$ ps). The spike signals that were separately observed in the IGM represent the center-bursts corresponding to the signal comb passed through the reference and sample paths, respectively. The "reference IGM" was applied to a computational real-time phase compensation using the Forman method,[15,16] which kept the observed interference waveforms constant during the experiment. This process artificially canceled the phase deviation between the signal and local combs, contributing the long-term coherent averaging far longer than the relative coherence time between the dual-comb sources.

The information about the sample was included in the "sample IGM," whose magnification is shown in Fig. 2(b). While scanning the pump delay time τ, the temporal waveform of the transmitted signal comb changes according to the dynamics of the optical properties of the sample, resulting in a tiny deformation of the IGM. In this experiment, we measured the response as the transient change of a normalized complex transmittance spectrum after the photo-excitation,

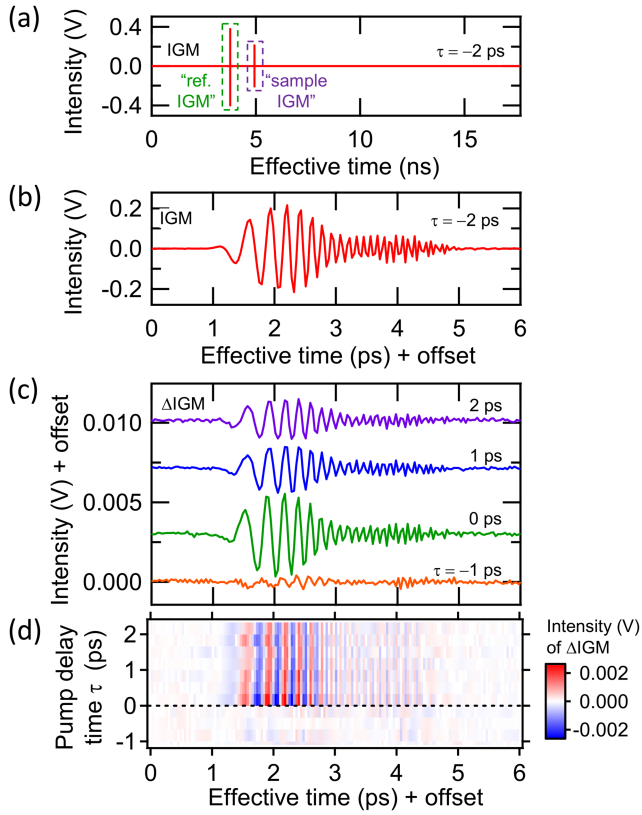

FIG. 2. (a) IGM observed with a pump delay time $\tau = -2$ ps. "Effective time" denotes the time interval between the sweeping pulses of the signal and local combs. (b) The magnification of the IGM at $\tau = -2$ ps around the "sample IGM." (c) Waveforms and (d) a two-dimensional color plot of transient optical responses of the IGM, $\Delta\mathrm{IGM}(\tau)$.

$\Delta\tilde{T}(\omega,\tau)/\tilde{T} = |\Delta T/T| \cdot \exp(i\Delta\phi)$, where ΔT denotes the transient change of the transmittance in amplitude (not in power), $\Delta\phi$ is that of the phase, and $\tilde{T}$ is the transmittance before the excitation; we adopted $\tilde{T}$ observed at $\tau = -2$ ps as the reference before the excitation in this experiment.

$\Delta\tilde{T}/\tilde{T}$ was obtained through the calculation of the ratio between the Fourier transformed spectra before ($\tau = -2$ ps) and after the excitation. Here, we measured $\Delta\tilde{T}/\tilde{T}$ every second by switching the pump delay time repeatedly and integrated the data. By adapting this acquisition scheme, we can cancel out the environmental phase drift caused by the slow change in the relative optical path length between the reference and sample paths, achieving a high phase resolution of a few milliradians (mrad) in this experiment. In the Fourier transform process, the part of the IGM around only the center burst was cut out, resulting in a GHz frequency resolution, which is sufficient to resolve the broadened spectra typically found in solid samples. This process contributes to reduce the real-time calculation cost. Here, it is noted that a higher frequency resolution can be obtained by tailoring the analytical conditions if necessary.

As a result, from the observed data, we obtained the dynamics of the IGM, $\Delta\mathrm{IGM}(\tau)$, as shown in Figs. 2(c) and 2(d). The temporal waveforms clearly show the ultrafast transient response induced by the excitation, whose amplitude changes on the order of 10^{-2} compared to the reference in Fig. 2(b).

In Figs. 3(a)–3(d), the transient complex transmittance spectra are summarized, exhibiting the dynamics of the amplitude and phase spectra, $\Delta T/T$ and $\Delta\phi$. In general, the amplitude and phase mainly reflect the responses of the absorption and the refractive index, respectively. The two-dimensional color plots shown in Figs. 3(a) and 3(b) display the transient changes in frequency vs pump delay time, τ. The spectra on the upper sides represent the cross sections of the color plots along the time axis. The waveforms shown in Figs. 3(c) and 3(d) are the temporal evolutions of $\Delta T/T$ and $\Delta\phi$ averaged over the frequency range of interest.

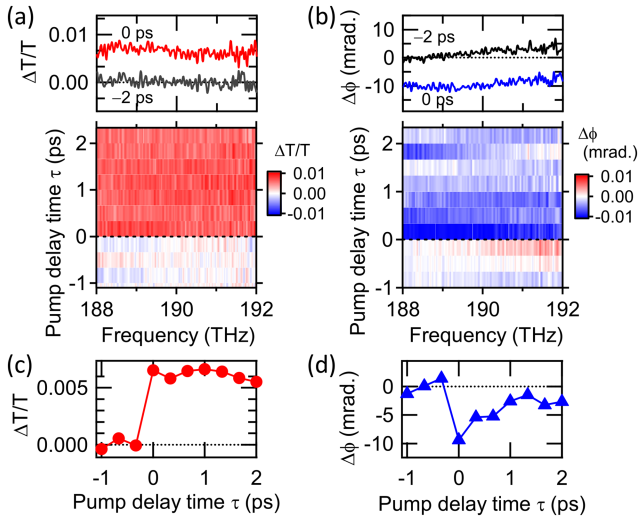

FIG. 3. Summary of the dynamics of the complex transmittance spectra, $\Delta\tilde{T}(\omega,\tau)/\tilde{T}$. (a) A two-dimensional color plot of the transient transmittance, $\Delta T/T$, and its cross sections along the time axis, and (b) those of the transient phase, $\Delta\phi$. The color denotes the positive (red) and negative (blue) changes. Temporal evolutions of (c) $\Delta T/T$ and (d) $\Delta\phi$ averaged over the frequency of interest.

In $\Delta T/T$ shown in Figs. 3(a) and 3(c), the spectrum has a flat structure, and $\Delta T/T$ steeply increases just after the excitation on the order of 10^{-3}, which is consistent to the results typically observed from the conventional pump–probe measurement for the semiconductor sample. On the other hand, $\Delta\phi$ shown in Figs. 3(b) and 3(d) has a similar flat spectrum and shows an approximately 10 mrad transient decrease just after the excitation. $\Delta\phi$ seems to have a relaxation component with a lifetime of about 1 ps, which is different from the behavior observed in $\Delta T/T$. We tentatively attributed this deviation to the induced phase modulation,[13] though the detailed assignment should be progressed in the future. In this way, the dynamics of the complex optical properties was successfully observed by the developed TR-DCS system. Both the amplitude and phase of the transmittance were separately characterized in the InGaAs sample.

In conclusion, we have achieved a new concept, time-resolved dual-comb spectroscopy (TR-DCS), which enables the direct observation of a transient complex optical spectrum by combining conventional DCS with the pump–probe method. As a demonstration, the femtosecond relaxation dynamics of a photo-excited InGaAs saturable absorber was investigated and both the transient amplitude and phase spectra of the complex transmittance were independently observed. In addition, the transient response of the interferogram, ΔIGM, was evaluated.

TR-DCS has great potential to be further progressed in the future. For example, by assuming an appropriate model around samples, we can deduce complex optical constants from the observed transient complex transmittance.[15] In characterizing samples with finer spectral components, TR-DCS will especially demonstrate its power with a high frequency resolution. Available frequency regions can be extended by using contemporary frequency conversion techniques. Furthermore, if an additional third comb oscillator, which has a slightly different repetition frequency, $f_{\mathrm{rep}} + \Delta f_{\mathrm{rep}}'$, is introduced as the pump comb, a further advanced two-dimensional spectroscopy, triple-comb spectroscopy, achieving much higher dynamic ranges without any mechanically moving part, should be realized. Moreover, one of the greatest advantages of the TR-DCS system is the ability to directly observe the electric fields of coherently controlled comb pulses, indicating a good compatibility with observations of coherent phenomena, such as the coherent Raman and four-wave mixing. We believe our proof-of-principle experiment will promote a pathway to coherent, highly accurate, and multi-dimensional pump–probe spectroscopy using the comb technology.

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[1] S. T. Cundiff and J. Ye, Rev. Mod. Phys. **75**, 325–342 (2003).
[2] F. Keilmann, C. Gohle, and R. Holzwarth, Opt. Lett. **29**, 1542–1544 (2004).
[3] I. Coddington, N. Newbury, and W. Swann, Optica **3**, 414–426 (2016).
[4] I. Coddington, W. C. Swann, and N. R. Newbury, Phys. Rev. A **82**, 043817 (2010).
[5] T. Ideguchi, S. Holzner, B. Bernhardt, G. Guelachvili, N. Picqué, and T. W. Hänsch, Nature **502**, 355 (2013).
[6] T. Ideguchi, B. Bernhardt, G. Guelachvili, T. W. Hänsch, and N. Picqué, Opt. Lett. **37**, 4498–4500 (2012).
[7] A. Hipke, S. A. Meek, T. Ideguchi, T. W. Hänsch, and N. Picqué, Phys. Rev. A **90**, 011805(R) (2014).
[8] A. Bartels, R. Cerma, C. Kistner, A. Thoma, F. Hudert, C. Janke, and T. Dekorsy, Rev. Sci. Instrum. **78**, 035107 (2007).
[9] A. Abbas, Y. Guillet, J.-M. Rampnoux, P. Rigail, E. Mottay, B. Audoin, and S. Dilhaire, Opt. Express **22**, 7831–7843 (2014).
[10] T. Yasui, E. Saneyoshi, and T. Araki, Appl. Phys. Lett. **87**, 061101 (2005).
[11] K. Minoshima, M. Taiji, and T. Kobayashi, Opt. Lett. **16**, 1683–1685 (1991).
[12] K. Minoshima, M. Taiji, A. Ueki, K. Miyano, and T. Kobayashi, Nonlinear Opt. **14**, 39–46 (1995).
[13] E. Tokunaga, A. Terasaki, and T. Kobayashi, Opt. Lett. **17**, 1131–1133 (1992).
[14] A. J. Fleisher, B. J. Bjork, T. Q. Bui, K. C. Cossel, M. Okumura, and J. Ye, J. Phys. Chem. Lett. **5**, 2241–2246 (2014).
[15] A. Asahara, A. Nishiyama, S. Yoshida, K. Kondo, Y. Nakajima, and K. Minoshima, Opt. Lett. **41**, 4971–4974 (2016).
[16] M. L. Forman, W. H. Steel, and G. A. Vanasse, J. Opt. Soc. Am. **56**, 59–63 (1966).