Spectrally Interleaved, Comb-Mode-Resolved, Dual-Terahertz-Comb Spectroscopy

Y.-D. Hsieh[1], Y. Iyonaga[1], Y. Sakaguchi[1], S. Yokoyama[1,2], H. Inaba[3,4], K. Minoshima[4, 5], F. Hindle[6], T. Araki[1], and T. Yasui[1, 4, 7,*]

[1]*Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan*
[2]*Micro Optics Co., Ltd, 2-20, Oe-Nakayama, Nishikyo, Kyoto 610-1104, Japan*
[3]*National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8563, Japan*
[4]*ERATO Intelligent Optical Synthesizer Project, JST, 1-5-1, Chofugaoka, Chofu, Tokyo 182-8585, Japan*
[5]*The University of Electro-Communications, 1-5-1, Chofugaoka, Chofu, Tokyo 182-8585, Japan*
[6]*Université du Littoral Côte d'Opale, 189A Av. Maurice Schumann, Dunkerque 59140, France*
[7]*The University of Tokushima, 2-1, Minami-Josanjima-cho, Tokushima, Tokushima 770-8506, Japan*
Author e-mail address: yasui.takeshi@tokushima-u.ac.jp

Abstract: We demonstrated combination of spectrally interleaved terahertz (THz) frequency comb with dual-comb spectroscopy, enabling us to achieve the spectral sampling equal to linewidth of the comb tooth in the low-pressure gas spectroscopy in THz region.

OCIS codes: (120.6200) Spectrometers and spectroscopic instrumentation; (300.6320) Spectroscopy, high-resolution; (300.6495) Spectroscopy, terahertz.

Optical frequency comb [1] and terahertz (THz) comb [2] have attracted attentions as a powerful tool for broadband spectroscopy because a series of the comb teeth can be used as frequency markers traceable to a microwave frequency standard for the broadband spectrum. Although the optical comb has been used in combination with Michelson-based Fourier-transform spectrometer (FTS) [3], dual-comb spectroscopy opens a new door for the optical-comb-based spectroscopy because it enables us to make full use of teeth in the optical comb [2,4]. However, too discrete distribution of the comb tooth limits the spectral sampling to the frequency spacing between the comb teeth although the linewidth of each tooth is sufficiently narrow. Here, we demonstrated combination of spectrally interleaved frequency comb with dual-comb spectroscopy in THz region by precise sweeping of frequency interval in dual optical combs [5]. Furthermore, the spectrally interleaved THz comb was effectively applied for high-precision spectroscopy of water vapor in low pressure.

To enhance the spectral sampling, frequency gap between the comb teeth has to be interleaved. THz comb is a harmonic frequency comb of a laser mode-locked frequency without carrier-envelope-offset frequency, and its frequency spacing is exactly equal to the mode-locked frequency [2]. Therefore, the absolute frequency of each comb teeth can be tuned by changing the mode-locked frequency. If incremental sweeping of the comb tooth is repeated at an interval equal to the tooth linewidth and the resulting all of the comb spectra are overlaid in the spectral domain as shown in Fig. 1, the frequency gaps between the comb teeth can be fully interleaved. In this way, the spectrally interleaved THz comb can be achieved. This situation is equivalent to continuous sweeping of a single-mode, narrow-linewidth CW-THz wave across a broadband THz spectral region. The resulting spectral sampling should be equal to the linewidth of the comb tooth.

To obtain the spectrum of THz comb, we used a time-window-extended, asynchronous-optical-sampling THz time-domain spectroscopy system consisting of dual mode-locked Er-fiber lasers (λ_c = 1550 nm, Δt = 50 fs, P_{mean} = 500 mW) and a THz optical setup. Detail of this system is given elsewhere [6]. The individual mode-locked frequencies of the two lasers (f_1 = 250,000,000 Hz and f_2 = 250,000,050 Hz) and the frequency offset between them ($\Delta f = f_2 - f_1$ = 50 Hz) were stabilized by two independent laser control systems referenced to a rubidium frequency standard. Furthermore, f_1 and f_2 can be respectively tuned within the frequency range of ±0.8 % by changing reference frequencies synthesized from the frequency standard. After acquiring the temporal waveform of 10 consecutive THz pulses with a fast digitizer (time window = 40 ns, sampling rate = 2 MS/s), we obtained the amplitude spectrum of THz comb by FT of the temporal waveform.

We evaluated the spectral resolution in the proposed method by measuring the rotational transition $1_{10} \leftarrow 1_{01}$ at 0.557 THz in water vapor enclosed in a low-pressure gas cell (length = 500 mm, diameter = 40 mm). To reduce the pressure broadening and the strong absorption, we diluted the water vapor at 10 Pa with a nitrogen gas at 320 Pa (expected pressure broadening linewidth = 23 MHz). Figure 2(a) shows the amplitude spectrum of the standard THz comb expanded around 0.557 THz [6]. The comb teeth had a frequency spacing of 250 MHz and a linewidth of 25 MHz. The frequency spacing was equal to the laser mode-locked frequency whereas the linewidth was consistent with the reciprocal of the temporal window. In this way, the amplitude spectrum without incremental sweeping of THz comb teeth did not indicate any spectral shape of the absorption line due to too coarse distribution of the comb teeth compared with the absorption linewidth of low-pressure water vapor. We next demonstrated incremental sweeping of the comb teeth across the absorption line at 0.557 THz by changing the mode-locked frequencies of both lasers. Incremental increase of f_1 and f_2 by 11,210 Hz were repeated ten times while keeping Δf at 50 Hz. The resulting overlaid spectra of comb modes are shown in Fig. 2(b). In this demonstration, a single shift of f_1 by 0.00448 % resulted in sweeping of the comb teeth by 10 % of its interval due to the large mode number of the THz comb (about 2,230 around 0.557 THz). Frequency gaps between comb teeth in Fig. 2(a) were fully interleaved. As a result, the sharp spectral dip was clearly appeared at the position of the water absorption line. Then, the absorption spectrum was obtained by extracting the peak amplitude of each comb teeth and normalization with a reference spectrum obtained under identical conditions, as shown in Fig. 2(c). The spectral linewidth was determined to be 24 MHz when a Lorentzian function was fitted to the spectral shape, indicated by the solid line in Fig. 2(c), which was consistent with the expected pressure broadening linewidth (= 23 MHz). Finally, we investigated pressure broadening of the same water line when the total pressure was changed. Black plots in Fig. 2(d) show the full-width at half-maximum (FWHM) of the observed absorption line with respect to the partial pressure of the water vapor, which was varied between 5 Pa and 160 Pa. For comparison, the expected characteristic of the pressure broadening is also indicated as a red line in Fig. 2(d) [7]. The experimental data were in good agreement with the expected curve down to 25 MHz, and then deviated from it. This result clearly indicated that the spectral resolution was enhanced down to the linewidth of the comb teeth (= 25 MHz) from their interval (= 250 MHz).

In summary, frequency gaps between THz comb teeth were successfully interleaved by simultaneous sweeping of the mode-locked frequencies in dual fiber lasers. The resulting spectral resolution was equal to linewidth of the comb tooth. The proposed method will be a powerful tool to simultaneously achieve high resolution, high accuracy, and broad spectral coverage in THz spectroscopy.

This work was supported by Collaborative Research Based on Industrial Demand from the Japan Science and Technology Agency, Japan. We also gratefully acknowledge financial support from the Renovation Center of Instruments for Science Education and Technology at Osaka University, Japan.

[1] Th. Udem, R. Holzwarth, and T. W. Hänsch, "Optical frequency metrology," Nature **416**, 233-237 (2002).
[2] T. Yasui, Y. Kabetani, E. Saneyoshi, S. Yokoyama, and T. Araki, "Terahertz frequency comb by multi-frequency-heterodyning photoconductive detection for high-accuracy, high-resolution terahertz spectroscopy", Appl. Phys. Lett. **88**, 241104 (2006).
[3] J. Mandon, G. Guelachvili, and N. Picqué, "Fourier transform spectroscopy with a laser frequency comb," Nature Photon. **3**, 99-102 (2009).
[4] F. Keilmann, C. Gohle, and R. Holzwarth, "Time-domain mid-infrared frequency-comb spectrometer," Opt. Lett. **29**, 1542-1544 (2004).
[5] T. Yasui, Y.-D. Hsieh, Y. Iyonaga, Y. Sakaguchi, S. Yokoyama, H. Inaba, K. Minoshima, F. Hindle, and T. Araki, "Spectrally interleaved, comb-mode-resolved spectroscopy using swept dual terahertz combs," Sci. Rep. **4**, *in press* (2014).
[6] Y.-D. Hsieh, Y. Iyonaga, Y. Sakaguchi, S. Yokoyama, H. Inaba, K. Minoshima, F. Hindle, Y. Takahashi, M. Yoshimura, Y. Mori, T. Araki, and T. Yasui, "Terahertz comb spectroscopy traceable to microwave frequency standard," IEEE Trans. Terahertz Sci. Tech. **3**, 322–330 (2013).
[7] T. Seta, H. Hoshina, Y. Kasai, I. Hosako, C. Otani, S. LoXowc, J. Urband, M. Ekström, P. Eriksson, and D. Murtaghd, "Pressure broadening coefficients of the water vapor lines at 556.936 and 752.033 GHz," J. Quantum Spectrosc. Radiat. Transfer **109**, 144–150 (2008).

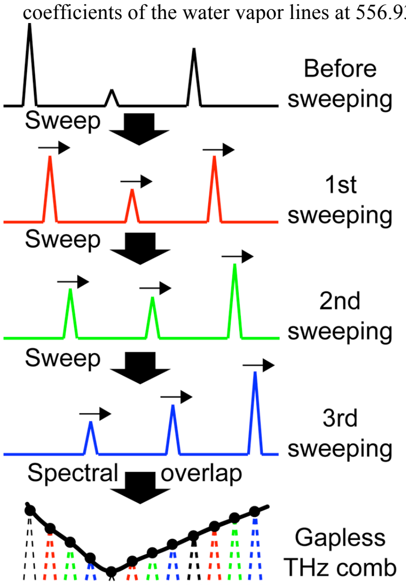

Fig. 1. Spectrally interleaved THz comb.

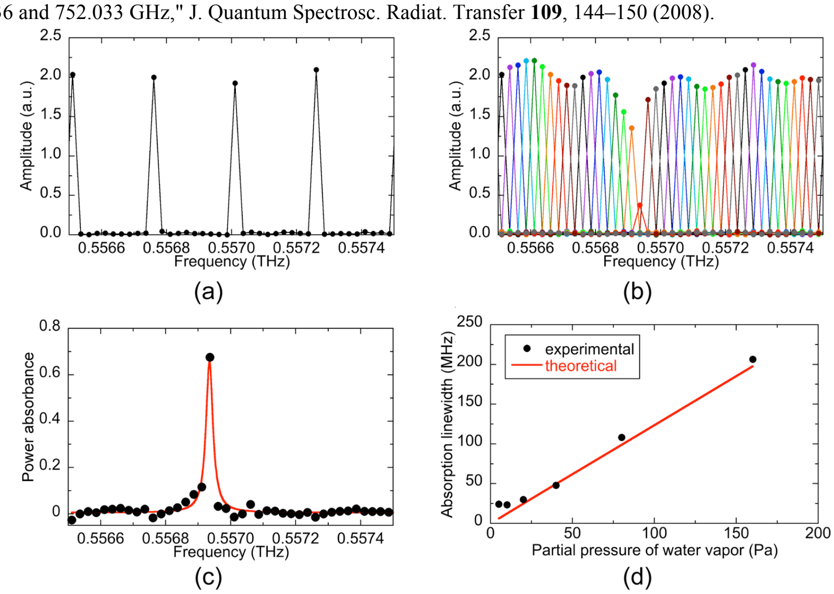

Fig. 2. Amplitude spectra of (a) standard THz comb and (b) spectrally interleaved THz comb around 0.557 THz after passing through low-pressure water vapor contained in the gas cell. (c) Absorption spectrum and (d) pressure broadening characteristics of the rotational transition $1_{10} \leftarrow 1_{01}$ in water vapor.