Terahertz Frequency Comb for High-accuracy, High-resolution Terahertz Spectroscopy

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Abstract: We report a terahertz frequency-comb technique for high-accuracy, high-resolution terahertz spectroscopy by combination of two mode-locked-frequency-stabilized femtosecond lasers and multi-frequency-heterodyning photoconductive detection.
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1. Introduction

A femtosecond mode-locked (fs-ML) laser generates a sequence of pulses that are essentially copies of the same pulse separated by an interval equal to the inverse of the mode-locked (ML) frequency. The highly stable fs-ML pulse train is synthesized by a series of frequency spikes regularly separated by the ML frequency in the optical frequency domain. Since this frequency-comb structure can be used as a precision ruler in the frequency domain, the fs-laser-based optical frequency comb has received a lot of interest as a powerful metrological tool capable of covering the visible to near-infrared region [1]. Recently, the concept of the frequency comb has been extended to the mid-infrared region by using a fs-ML laser in combination with the nonlinear difference frequency generation process [2], and effectively applied to a frequency-comb-based Fourier-transform infrared spectrometer based on a multi-frequency-heterodyning interferometeric technique [3, 4]. On the other hand, a THz electromagnetic pulse, radiating from a photoconductive antenna or nonlinear optical crystal triggered by a fs-ML laser, is also composed of a regular comb of sharp lines in the THz-frequency domain. This so-called THz frequency comb possesses attractive features for high-precision THz spectroscopy, namely excellent accuracy and stability, broadband selectivity, high spectral purity, and exact multiplication. Furthermore, since the THz frequency comb is extended to the THz region without any frequency offset, it is possible to achieve absolute calibration without the use of a standard laser or a standard material. If the THz comb can be used as a frequency ruler in THz spectroscopy, high-accuracy, high-resolution THz spectroscopy will be achieved. In this presentation, we propose a THz frequency-comb technique by combined use of two ML-frequency-stabilized fs-ML lasers and multi-frequency-heterodyning photoconductive detection [5].

2. Principle and experimental setup

Let us consider THz generation from a photoconductive emitter excited by a fs-ML laser (pump laser; ML freq. = f_1) and THz detection using a photoconductive detector gated by another fs-ML laser (probe laser; ML freq. = f_2) as shown in Fig. 1. Figure 2 illustrates spectral behaviors in (a) optical, (b) THz, and (c) radio-frequency (RF) regions. In the frequency domain, since the photoconductive generation of the THz pulse can be considered as an ultra-wideband demodulation of an optical frequency comb, the frequency comb is down-converted to the THz region without any change to the frequency spacing. The resulting THz comb is a harmonic frequency comb without a frequency offset, composed of a fundamental component (freq. = f_1) and a series of harmonic components (freq. = $2f_1$, $3f_1$, •••••••, nf_1) of a ML frequency. Next, we consider what happens when the ML frequency of the probe laser ($f_2=f_1+\Delta f$) is slightly detuned from that of the pump laser (f_1) by a certain frequency offset (Δf). Instantaneous photoconductive gating by the probe laser induces a photocurrent frequency comb having a different frequency spacing [freq. = f_2, $2f_2$, $3f_2$, •••••••, nf_2 = $(f_1+\Delta f)$, $2(f_1+\Delta f)$, $3(f_1+\Delta f)$, •••••••, $n(f_1+\Delta f)$] in the photoconductive detector, which is also existed in THz-frequency region. Under this condition, it is possible to

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detect the THz pulse as a result of the photoconductive process occurring between the THz and photocurrent frequency combs, giving rise to the multi-frequency-heterodyning effect. This results in the generation of a secondary frequency comb in the RF region, termed the RF electric comb (freq. = Δf, 2Δf, 3Δf, •••••••, nΔf). Since the RF comb is a replica of the THz comb only downscaled by $f_1/\Delta f$ in frequency, one can utilize the THz frequency comb easily via direct observation of the RF frequency comb using an RF spectrum analyzer and calibration of the frequency scale.

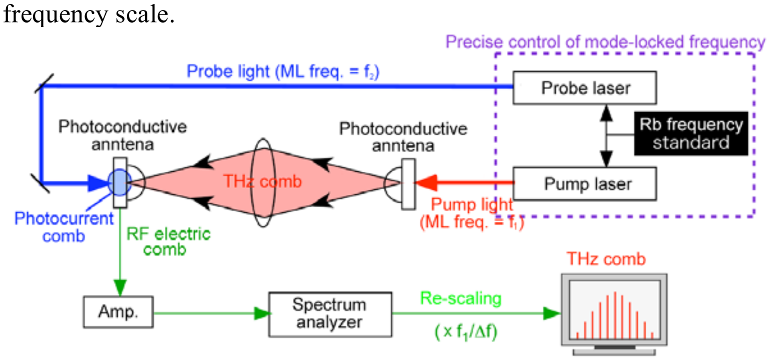

Fig. 1 Experimental setup.

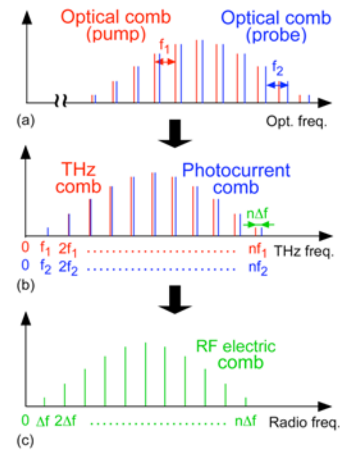

Fig.2 Principle.

3. Results

Figure 3 shows an amplitude spectrum of the RF frequency comb obtained using the proposed method [measurement time = 10 sec, resolution bandwidth = 10 kHz], in which the upper horizontal axis gives the frequency scale in the RF spectrum analyzer. The actual frequency values in the THz frequency-comb spectrum are scaled according to the lower horizontal axis by using a reciprocal of the frequency downscale factor. Considering a spectral range of 0.3 THz and a frequency spacing of 81.8 MHz, the THz spectrum is composed of 3,667 comb modes. The inset of Fig. 3 shows an expansion of the spectral region of 0.0492 ~ 0.0498 THz (measurement time = 0.5 sec, resolution bandwidth = 10 Hz). One can clearly see 7 comb-mode lines having a frequency spacing of 81.8 MHz and a width of 11 MHz; these can be used as divisions in a THz frequency-comb ruler with a frequency resolution of 81.8 MHz. The accuracy of the observed THz frequency comb is determined by stability of frequency spacing and frequency downscale factor. We can conclude the accuracy in the proposed THz-comb spectroscopy to be 2.5×10^{-7} from stability of f_1 [$= (1.6*10^{-3})/81{,}800{,}000 = 2.0\times10^{-11}$] and that of Δf [$= (2.5*10^{-5})/100 = 2.5\times10^{-7}$].

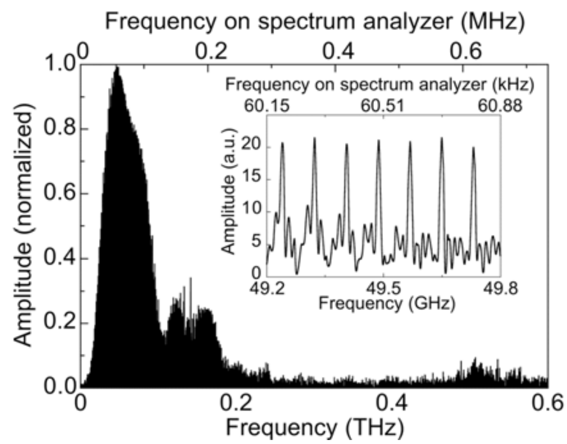

Fig.3 Amplitude spectra of THz frequency comb.

4. Summary

We demonstrated a THz frequency-comb technique based on multi-frequency-heterodyning photoconductive detection. The proposed method will be a powerful spectroscopic and metrological tool in THz frequency region.

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