High-resolution terahertz spectroscopy with a single tunable frequency comb

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Abstract: We report an improvement of three orders of magnitude in the spectral resolution of a recently proposed single-comb terahertz spectroscopy [Opt. Lett. **39**, 5669 (2014)]. The improvement is achieved by using a femtosecond optical pulse train with a tunable repetition rate. Terahertz comb with tunable spectral line spacing generated by the train is detected via nonlinear mixing with a harmonic of a CW signal from a microwave frequency synthesizer. By applying this technique to the low-pressure gas spectroscopy, we achieved a 100 kHz spectral resolution in measuring separate absorption lines of the rotational manifold of fluoroform (CF_3H).

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

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1. Introduction

Terahertz spectroscopy based on frequency combs has been developed in recent years [1–6] as an extension of the preceding techniques that revolutionized high-precision metrology and spectroscopy in the visible and mid-infrared spectral regions [7–9]. In conventional terahertz frequency comb spectroscopy, dual optical combs are used [1–6,9]. The first optical comb is down-converted to the terahertz region by using a biased photoconductive antenna (PCA) and generates a comb of terahertz waves with the same spectral line spacing as in the optical comb. After transmission through a sample, the terahertz comb carries the spectroscopic information about the sample encoded in the amplitudes of the comb spectral components. To read this information, the terahertz comb is sent to another (non-biased) PCA gated by the second optical comb with slightly different spectral line spacing. The second optical comb generates a terahertz comb of photocarriers in the PCA. Since the photocurrent in the PCA-detector is proportional to the product of the electromagnetic and photocarrier terahertz combs, its spectrum is a replica of the electromagnetic terahertz comb downscaled into the radio frequencies (RF). By measuring this spectrum with an RF spectrum analyzer, one can visualize the terahertz comb.

Implementation of the conventional dual-THz-comb technique requires a dual femtosecond laser system for generating two synchronized optical combs. Recently, terahertz spectroscopy with a single optical comb has been introduced and experimentally demonstrated [10]. In this technique, the optical comb from a single femtosecond laser is used to produce an electromagnetic terahertz comb. For detecting the terahertz comb, it is heterodyned to the UHF frequency range via nonlinear mixing in a superlattice (SL) diode with a harmonic of a sinusoidal EHF signal from a frequency synthesizer. The spectral resolution of such single-THz-comb spectroscopy is determined by the spectral line spacing of the terahertz comb, which equals the repetition rate of the pump laser (100 MHz in [10]). This exceeds the resolution of the standard terahertz time-domain spectroscopy (THz-TDS) [11] by the order of magnitude. The spectral coverage of the technique is determined by the nonlinear properties of the mixer and was as large as several hundred gigahertz in [10].

In the present paper, we propose and demonstrate experimentally a method for improving the spectral resolution of the single-THz-comb spectroscopy by three orders of magnitude. To achieve the improvement, we use a commercially available femtosecond fiber laser with a variable cavity length as a source of the optical comb. The repetition rate (100 MHz) of the laser, and therefore the spectral line spacing of the terahertz comb generated with this laser, can be varied in the frequency range of 330 kHz with a 4 Hz step by changing the cavity length with a motorized translational stage. Correspondingly, the frequency of the nth component of the terahertz comb can be tuned with an n times larger step. For example, the

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frequency (400 GHz) of the n = 4000 component can be tuned in a 1.32 GHz range by a 16 kHz step and thus sweep the spectral line of interest of a sample with the 16 kHz accuracy. In our experiment, however, this accuracy was excessive due to spectral impurity of the frequency synthesizer (~20 kHz) and was reduced to ~100 kHz by a corresponding increase of the tuning step. By applying this technique to the low-pressure gas spectroscopy, we achieved a 100 kHz spectral resolution in measuring separate absorption lines of the rotational manifold of fluoroform (CF_3H).

2. Experimental setup and principle of operation

A schematic diagram of the experimental setup is shown in Fig. 1. As a source of the optical comb we use a mode-locked femtosecond Er-doped fiber laser with 1.56 μm central wavelength, 65 fs pulse duration, 30 mW average power, and 100 MHz repetition rate (Menlo Systems GmbH, model C-fiber). The repetition rate of the laser can be tuned in the frequency range of 330 kHz by 4 Hz steps. It is achieved by changing the cavity length with a stepper motor (within each step the repetition rate can be additionally tuned with a 1 μHz accuracy by using a piezo actuator). By using PCA (Menlo Systems GmbH, model Tera 15-SL25, 20 V bias voltage), the optical comb is down-converted into a comb of terahertz waves. The terahertz beam is collimated by a TPX lens, then transmitted through a low-pressure gas cell with Brewster angle input/output quartz windows of 2 mm thickness, and finally focused onto the entrance aperture of a horn antenna by the second TPX lens. The low-frequency components of the terahertz signal are filtered out by waveguide 1 with a 187.5 GHz cutoff frequency. The higher frequency components are fed to a SL diode, which is used as a nonlinear mixer. Simultaneously the diode is fed (through waveguide 2) by a CW signal from a frequency synthesizer with a 78-178 GHz tuning range and 20 mW output power (the power of the CW signal on SL can be regulated by an attenuator). The SL diode used in the experiment contains a 112-nm-thick GaAs/AlAs superlattice with 18 periods each of which contains 18 monolayers of GaAs and 4 monolayers of AlAs (see details in [12,13]). The diode is characterized by an N-type nonlinear current-voltage curve, typical for superlattices [14]. Due to the nonlinearity of the SL diode, the current flowing in the diode contains harmonics of the CW signal, any one of which can be used as a local oscillator to downconvert the terahertz signal into RF, as in a standard heterodyne scheme [14]. The downconversion is performed by difference-frequency generation between the terahertz signal and a harmonic of the CW signal in the SL diode. As a result, the mixer produces multiple beat signals at RF. The beat signals which fall into the bandwidth of the amplifier ~280-530 MHz are amplified by 53 dB and observed with an RF spectrum analyzer (Agilent Technologies E4407B).

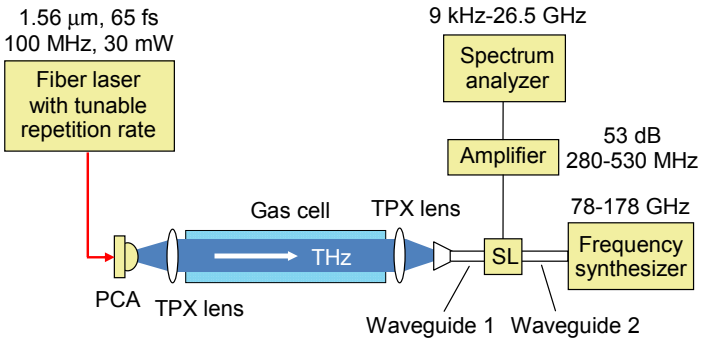

Fig. 1. Experimental setup.

The principle of the operation is illustrated by Fig. 2. The terahertz comb with the spectral line spacing equal to the repetition rate f_r of the pump laser is mixed with the harmonics of the CW signal carrier f_s (for quadratic nonlinear conductivity of SL, only even harmonics are

#225537 - $15.00 USD Received 22 Oct 2014; revised 15 Dec 2014; accepted 15 Dec 2014; published 22 Dec 2014

generated), see Figs. 2(a) and 2(b). Mixing produces multiple beat signals at the frequencies $|nf_r - mf_s|$, where n and m are integers, i.e., RF comb. For a fixed m, there are two sets of the RF comb components with n corresponding to $nf_r - mf_s > 0$ and $nf_r - mf_s < 0$ [Fig. 2(c)]. For spectroscopic measurements, we choose the comb component nf_r, which is closest to the sample's spectral line of interest. Then we tune the harmonic mf_s to obtain the beat signal with nf_r at the frequency f_b in the center of the amplifier bandwidth [Fig. 2(b)]. By changing the repetition rate f_r with a Δf_r step, we sweep the spectral interval of interest with a $n\Delta f_r$ step. To keep the beat frequency f_b constant during the sweeping, we tune simultaneously the harmonic mf_s by varying the frequency of the CW signal with a $(n/m)\Delta f_r$ step [Fig. 2(b)]. Keeping f_b constant allows us to exclude the effect of inhomogeneity of the frequency response of the amplifier on the acquired spectroscopic information. (In the experiment, we fixed f_b at the central frequency of the amplifier bandwidth, f_b = 420 MHz.) The spectroscopic information is retrieved from the power A_b of the RF signal at the frequency f_b by using the spectrum analyzer [Fig. 2(c)].

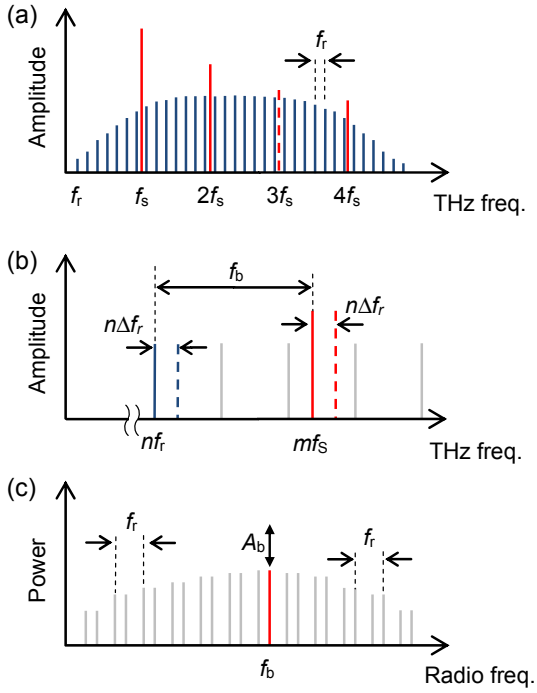

Fig. 2. a) Terahertz comb (cyan) and harmonics of the CW signal (red). b) Enlarged part of (a). The comb components after tuning are shown in dashed lines. c) RF comb with two sets of components. Amplitude A_b of the component at f_b changes with tuning f_r and f_s.

3. Spectroscopy of fluoroform

To assess the capacity of the technique to resolve fine spectral features, the absorption profile of the rotational manifold of the $J - J' = 13 - 14$ transition of fluoroform (CF_3H) near 0.2897 THz at a pressure of 100 mTorr was measured. At such a low pressure the fourteen K-components of the manifold are well separated, as can be seen from the theoretical transmission spectrum in Fig. 3(a). (K is the quantum number that characterizes the projection of the total angular momentum, defined by the quantum number J, onto the symmetry axis of the molecule.) To perform the measurements, a quartz gas cell (a 25-mm diameter and 1000-mm length) with CF_3H was placed between the TPX lenses (Fig. 1). We measured the amplitude of the n = 2896 component of the terahertz comb, which was downconverted to f_b =

#225537 - $15.00 USD Received 22 Oct 2014; revised 15 Dec 2014; accepted 15 Dec 2014; published 22 Dec 2014

420 MHz by mixing with the second (m = 2) harmonic of the CW signal: $2f_s - 2896f_r = 420$ MHz.

At first, we measured the transmission spectrum in a wide frequency interval 289.64-289.73 GHz with a 500 kHz step by increasing f_r (with a 500/2896 kHz step) and tuning f_s simultaneously to keep f_b constant. The measurement time for each data point was about 1 min (~180 min in total). The results are shown in Fig. 3(b). The molecular absorption is observed on a non-linear baseline, which is typical for the spectrometers with such geometry as in Fig. 1 and results from uncontrolled reflection of radiation from the spectrometer elements. Almost all absorption lines of the manifold are well resolved except for a few close lines in the low-frequency part of the spectrum. The central frequencies of the resolved lines agree well with the theoretical frequencies in Fig. 3(a). The theoretical spectrum in Fig. 3(a) was calculated using the routine method for symmetric-top molecules based on Eq. (3-51) in [15] and Eq. (4) in [17] for the spectral line intensities and center frequencies, respectively. The rotational, centrifugal, and other high order efficient constants for the CF_3H molecule were taken from [16, 17] and the collisional broadening parameter 10 MHz/Torr (half-linewidth) was obtained by extrapolating the experimental J-dependence of this parameter [18] to large numbers J. The Doppler broadening (~0.2 MHz) is negligible for 100 mTorr.

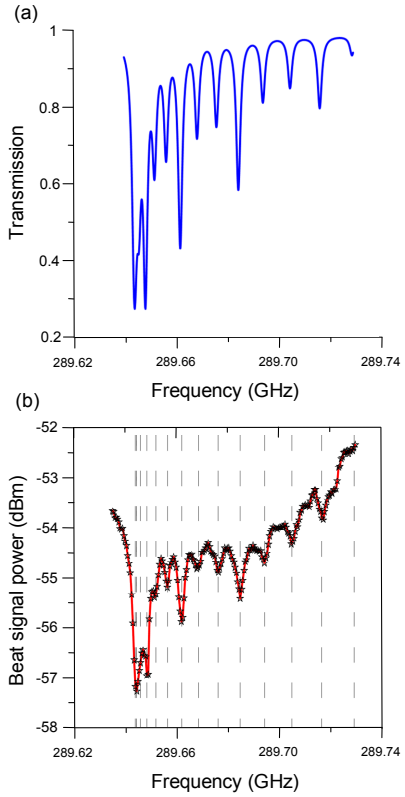

Fig. 3. Transmission spectrum of CF_3H at 100 mTorr near 289.7 GHz: a) theoretical and b) experimental. In (b), vertical dashed lines show the theoretical frequencies from (a).

Next, we measured the profile of one of the manifold lines (with K = 6) around 289.662 GHz (Fig. 4). To evaluate the center frequency of the line and spectral linewidth, we fitted a Lorentzian function to the measured profile. Using a Lorentzian function rather than the generally used Voigt function is justified by the domination of the pressure broadening over the Doppler one at 120 mTorr. The obtained center frequency 289.66205 GHz and linewidth 3.02 MHz (FWHM) were compared with corresponding theoretical values 289.66215 GHz

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and 2.4 MHz calculated using the same approach and parameters as for calculating the spectrum in Fig. 3(a). The center frequency agrees well with the theoretical value. The somewhat larger experimental linewidth can be explained by neglecting the effect of the other manifold lines and the baseline on the theoretical linewidth.

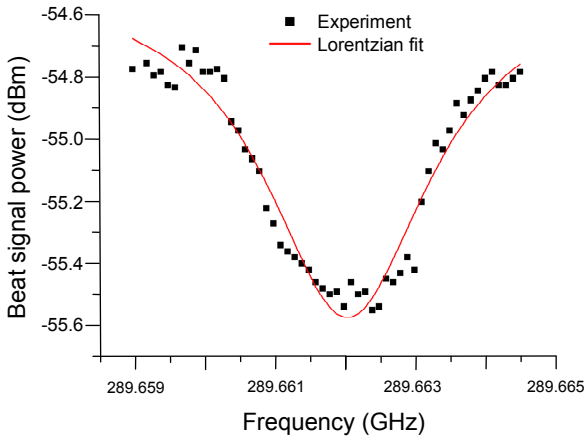

Fig. 4. The spectral line with $K = 6$ of CF_3H at 120 mTorr.

4. Conclusion

We demonstrated single-THz-comb spectroscopy, based on electronic heterodyning of a photonically generated comb, with a spectral resolution of ~100 kHz, i.e., three orders of magnitude higher than in the first demonstration of the technique [10]. The improvement is achieved by using a pump femtosecond laser with a tunable repetition rate. The spectral resolution is defined by the tuning step of the laser and the spectral purity of the microwave frequency synthesizer, which is used for heterodyne detection of the terahertz comb. The advantage of the proposed technique over the standard dual-THz-comb spectroscopy is that it does not require a complex double-laser system but still has a high spectral resolution and wide spectral coverage. In principle, the spectral coverage of the technique is only limited by the bandwidth of the femtosecond laser pulse, i.e., coincides with the bandwidth of THz-TDS. Technically, the coverage is also affected by the properties of the nonlinear mixer. The mixers based on GaAs/AlAs quantum semiconductor superlattices can operate at frequencies up to 8 THz [19]. For our experimental setup, where a relatively low frequency superlattice diode is used, the coverage is as large as several hundred gigahertz – from ~200 to ~500 GHz (see Fig. 5 in [10]). In the vicinity of any frequency from this interval, using the technique presented above allows one to perform precise spectroscopic measurements with ~100-kHz resolution. The spectral coverage can be extended to the higher frequencies by using a higher frequency superlattice mixer.

If compared with more traditional methods of high-precision terahertz spectroscopy, such as tunable far-infrared spectroscopy [20, 21] or far-infrared gas laser sideband spectroscopy [22], the proposed technique is free from their inherent limitations including a very low power, non-continuous spectral coverage, and the bulky and complex instrumentation. We demonstrated that the proposed technique has potential for high-precision spectroscopy of gas molecules.

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