Low-Pressure Gas Spectroscopy Using Terahertz Frequency Synthesizer Traceable to Microwave Frequency Standard via Dual Optical Comb

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Abstract—We constructed a widely and continuously tunable terahertz frequency synthesizer traceable to a microwave frequency standard. Photomixing of two optical frequency synthesizers, linked to the frequency standard via dual optical frequency combs, makes this THz synthesizer traceable to the frequency standard. To demonstrate the potential of wide and continuous tunability in the THz synthesizer, we tuned its output frequency by 120 GHz continuously around 0.65 THz by using a uni-traveling-carrier photodiode as a photomixer and applied it for precise THz spectroscopy of low-pressure molecular gas. This THz synthesizer will be a powerful tool for broadband, high-precision THz spectroscopy.

I. INTRODUCTION

Accurate, stable, and tunable single-frequency signal generator in THz region, namely THz synthesizer, is an important technique in frequency metrology, precise spectroscopy, local oscillator for heterodyne receiver, and carrier wave generation for broadband wireless communications. One promising method for widely tunable CW-THz synthesizers is photomixing of two CW near-infrared lasers of adjacent wavelengths with a photomixer. However, if the two CW lasers are operated in the free-running mode, it is difficult to generate the accurate and stable frequency in the CW-THz wave. Recently, accurate, stable, phase-locked CW-THz wave has been discretely generated at four different frequencies (0.30, 0.56, 0.84, and 1.1 THz) by photomixing of two CW lasers locked to a single optical comb based on a mode-locked Ti:Sapphire laser [1]. Furthermore, the output frequency was tuned continuously by scanning the frequency interval of the optical comb while locking the CW lasers to the comb; however, the range of continuous tuning was limited to 11 MHz [2]. In the case where the two CW lasers share the same optical comb, when scanning the frequency interval of the comb, the optical frequencies of the two CW lasers change simultaneously. This common-mode change cancels most of the optical frequency change in the two CW lasers. As a result, the continuous tuning range of the CW-THz wave is much smaller than that of the optical frequency in the CW lasers. To increase the continuous tuning range of the CW-THz wave while maintaining high frequency accuracy and stability, a THz synthesizer has been achieved by photomixing of an accurately tunable CW laser and a tightly fixed CW laser in the optical frequency region, phase-locked to dual optical combs [3,4]. However, the generation frequency of the CW-THz wave has remained within the F-band of 90-140 GHz. In this paper, we achieved a higher-frequency, more broadband THz synthesizer, which can operate from 0.2 to 1.8 THz, by using a uni-traveling-carrier photodiode (UTC-PD) for a broadband photomixer, and applied it for low-pressure THz gas spectroscopy.

II. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. We prepared two CW lasers operating at a wavelength of 1550 nm for photomixing (CWL1 and CWL2: external cavity laser diode) and optical frequency difference between them is set to be around 0.6 to 0.7 THz by using an optical wavemeter. CWL1 and CWL2 were respectively phase-locked to two independent optical combs of commercial mode-locked Er-doped fiber combs operating at a center wavelength of 1550 nm (FC1500-250s, Menlo Systems; COMB1 and COMB2). The frequency interval (frep1, frep2) and carrier-envelop offset frequency (fceo1, fceo2) of COMB1 and COMB2 were phase-locked to the rubidium frequency standard. Combination of CWL1 and COMB1 was used as a fixed optical frequency synthesizer (OFS1, ffofs1 = fceo1 + frep1 + fbeat1; frenp1 = 250MHz, fceo1 = -20 MHz, fbeat1 = 773099, frep1 = 30 MHz) whereas another combination of CWL2 and COMB2 was used as a tunable optical frequency synthesizer (OFS2, ffofs2 = fceo2 + frep2 + fbeat2; frenp2 = 250MHz, fceo2 = -20 MHz, fbeat2 = 775499, frep2 = 30 MHz). The ffofs2 could be continuously tuned over 100 GHz without losing phase-locking of CWL2 to COMB2 by varying the repetition frequency over 128.949 kHz with a variable optical delay line module in COMB2. Outputs of OFS1 and OFS2 are combined with a fiber coupler, amplified, and then photomixed by a broadband uni-traveling-carrier photodiode (UTC-PD, NTT Electronic, available frequency range = 0.2–1.8 THz). Photocurrent of the UTC-PD is set to be 6 mA (estimated output power = 3 μW for 0.65 THz) by adjusting output power of CWL1 and CWL2. The generated CW-THz wave with a linewidth of 500 kHz passes through a low-pressure gas cell (diameter = 40 mm, length = 400 mm) and is measured by a pyrometer and a lock-in amplifier.
III. RESULTS

Acetonitrile (CH$_3$CN) is a very abundant species in the interstellar medium and is an ideal observational probe of the kinetic temperature and density of interstellar clouds. Furthermore, it is closely related with atmospheric pollution because of volatile organic compound. Therefore, there is a considerable need for probing this molecule. We performed gas-phase spectroscopy of this molecule to evaluate a potential of the proposed system for gas analysis. Since CH$_3$CN is a symmetric top molecule with a rotational constant $B$ of 9.2 GHz, it displays two features: a series of grouped rotational transitions regularly spaced by $2B (= 18.4 \text{ GHz})$, and hyperfine structure of rotational transitions into each transition group determined by the centrifugal distortion constant $D_{JK}$. Figure 2(a) shows an absorption spectrum of CH$_3$CN at a pressure of 20 Pa when scanning the CW-THz frequency from 0.6 to 0.72 THz continuously. Seven groups from $J = 32$ to $J = 38$ was clearly confirmed at intervals of 18.4 GHz within this frequency range. Next, we expanded the frequency range around 0.62485 THz to observe the hyperfine structure into a single group ($J = 33$), as shown in Fig. 2(b). In comparison with literature values reported in the JPL database [5], we successfully assigned lines $K = 0$ to 3 within a frequency discrepancy of -0.14 ±0.3 MHz (mean ± standard deviation for 4 absorption lines). These results clearly indicated a high potential of the proposed THz synthesizer for low-pressure gas-phase spectroscopy.

IV. SUMMARY

The precise THz synthesizer based on dual optical combs enables us to perform the precise THz spectroscopy secured by a microwave frequency standard.

REFERENCES