Development of a Carbon-Monoxide Rotational-Transition-Stabilized 3.1THz Quantum Cascade Laser for Terahertz Frequency Standard

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Abstract—We have stabilized the frequency of a 3.1 THz quantum cascade laser (QCL) to the rotational transition $J = 27 \leftarrow 26$ of carbon-monoxide (CO). The achieved frequency stability was 5×10^{-9} in the Allan standard deviation at 10s averaging time. The absolute frequency of the THz-QCL was determined to be 3097 909.4 (0.1) MHz, which agrees to the previous report within the uncertainty [1]. This CO-stabilized THz-QCL has a potential to be served as an unprecedented frequency standard in THz region.

I. INTRODUCTION

E STABLISHMENT of frequency standard in the THz domain (0.1 THz \sim 10 THz) is requested to allocate the THz spectrum among a wide range of users in ultra-highspeed communication, security, astronomy etc. National Institute of Information and Communications Technology (NICT) is developing a new THz frequency standard, which will lie in the so-called THz gap (Fig. 1). A THz-QCL stabilized to the clock transition of molecules can become a such THz standard with the sufficient accuracy for present THz users.



Fig. 1. Summary of list of radiations (LoR) and secondary representations of the second.

Carbon-monoxide (CO) is a diatomic molecule having many absorption lines of pure rotational transition between 0.1 THz and 4 THz with approximately 0.1 THz spacing. Their Zeeman and Stark shifts caused by the external electromagnetic field are intrinsically small because of no nuclear spin and small electric dipole moment. Moreover, both shifts can be calculated relatively easily, since CO has a simpler energy structure than asymmetric-top molecules such as H₂O and CH₃OH. The expected uncertainty of systematic frequency shift of the rotational transition $J = 27 \leftarrow 26$ was estimated to be less than 10 kHz by means of the coefficients of major shifts as summarized in Table I. Thus, this rotational transition is suited as an absolute frequency reference of THz-QCL. We have stabilized the frequency of a 3.1THz-QCL with 2 mW output power to the rotational transition $J = 27 \leftarrow 26$ of CO molecules.

TABLE I COEFFICIENTS OF SYSTEMATIC FREQUENCY SHIFT OF $^{12}\mathrm{C}^{16}\mathrm{O}$ rotational transition $J=27\leftarrow26$

| Transition frequency | 3 097 909.360 (0.017) MHz [1] |
|----------------------|---|
| Stark shift | $0.1\mathrm{mHz}/(\mathrm{V/cm})^2$ |
| Zeeman shift | $\sim mHz/G$ ($\Delta M = 0$) |
| | $\sim \rm kHz/G$ ($\Delta M = \pm 1$) |
| Collisional shift | $\leq -30 \text{kHz/Torr}$ [3], [4] |

II. EXPERIMENT AND RESULTS



Fig. 2. Experimental setup for THz-QCL frequency stabilization.

As the THz-QCL obtained was oscillating at several GHz higher frequency than the target one, the modified gas condensation method was employed to compensate this frequency discrepancy [2]; CO_2 gas was used instead of N_2 gas due to the deposition property matched with the operational condition of our THz-QCL.

Figure 2 illustrates the experimental setup for the frequencystabilized THz-QCL. The frequency-modulation spectroscopy technique was employed to extract the feedback control signal of THz-QCL. The THz light was modulated by a 33.7 kHz sinusoidal signal and introduced into a 30 cm CO-gas cell with 2 Torr pressure. The light transmitted through the cell was detected by a Fermi-level managed barrier diode [5], and its output voltage signal was demodulated to obtain a firstderivative signal of linear absorption spectrum (Fig. 3). The spectrum linewidth was about 15MHz, which was restricted by the Doppler and collisional broadening. The first-derivative signal was fed back to the current of the THz-QCL via a resistive summer to engage the frequency lock.



Fig. 3. First-derivative signal of rotational-transition spectrum of CO molecule. Inset: Linear absorption spectrum of $J = 27 \leftarrow 26$ transition.

The instability of the THz-QCL was measured by a semiconductor-superlattice harmonic mixer (SLHM) [6], [7] whose local oscillator (LO) was referenced to Japan Standard Time. The heterodyne beat signal between the THz-QCL and the LO harmonics produced in the SLHM had a SNR of 20dB in a 500kHz resolution bandwidth. The frequency jitter and drift of the THz-QCL was successfully suppressed, and then the Allan standard deviation was reached to 10^{-9} -level around 10 s averaging time as plottted in Fig. 4. Its absolute frequency was determined to be 3 097 909.4 (0.1) MHz, which is good agreement with the previous value [1].



Fig. 4. Allan standard deviation of CO-stabilized THz-QCL. Inset: RF spectra of heterodyne beat signal of frequency-locked (red) and free running (blue).

REFERENCES

- T.D. Varberg and K.M. Evenson, "Accurate far-infrared rotational frequencies of carbon monoxide," Astro-phys. J., 385, 763 (1992).
- [2] D. Turčinková, M.I. Amanti, F. Castellano, M. Beck, and J. Faist, "Continuous tuning of terahertz distributed feedback quantum cascade laser by gas condensation and dielectric deposition," Appl. Phys. Lett. 102, 181113 (2013).
- [3] S.P. Belov, M.Yu. Tretyakov and R.D. Suenram, Astro-phys. J., "Improved laboratory rest frequency measurements and pressure shift and broadening parameters for the J = 21 and J = 32 rotational transitions of CO," **393**, 848,(1992).
- [4] M. Fabian, I. Morino and K.M.T. Yamada, J. Mol. Spectrosc., "Absolute intensity measurement of the CO $J = 1 \leftarrow 0$ transition at 115 GHz," **185**, 422 (1997).

- [5] H. Ito and T. Ishibashi, Electron. Lett., "Low-noise heterodyne detection of terahertz waves at room temperature using zero-biased Fermi-level managed barrier diode," 54, 1080 (2018).
- [6] D.G. Paveliev, Yu.I. Koshurinov, V.P. Koshelets, A.N. Panin, V.L. Vaks, "Superlattice harmonic mixer for submillimeter frequency synthesis," Conference Digest of the 12th Int. Conf. on THz Electronics, Universität Karlsruhe (2004), p.327.
- [7] K.F. Renk, A. Rogl, B.I. Stahl, A. Meier, Yu.I. Koschurinov, D.G. Pavel'ev, V. Ustinov, A. Zhukov, N. Maleev, and A. Vasilyev, "Semiconductor-superlattice frequency mixer for detection of submillimter waves," Int. J. Infrared Milli. Waves, 27, 373 (2006).