Millimeter-Wave Oscillator Disciplined by Molecular Rotational Spectroscopy

James Greenberg and Antoine Rolland
Boulder Research Laboratory
IMRA America, Inc.
1551 S. Sunset Street, Longmont, CO, USA
jgreenbe@imra.com

Martin E. Fermann
IMRA America, Inc.
1044 Woodridge Avenue, Ann Arbor MI, USA

***Summary*— We present a millimeter-wave oscillator locked to a rotational transition of gaseous nitrous oxide (N_2O). Millimeter-wave radiation is generated by an optical heterodyne of two diode lasers on a uni-traveling-carrier photodiode (UTC-PD). We perform wavelength modulation spectroscopy at 301.44 GHz to discipline the frequency difference between the diode lasers and thus, lock the millimeter-wave radiation to the molecular rotational line. We measure the frequency of the oscillator via electro-optic (EO) frequency down-conversion, referenced to a Rubidium (Rb) clock. This results in an out-of-loop fractional frequency stability of the oscillator at 10^{-11} at one second averaging time.**

Keywords—millimeter-wave oscillator; wavelength modulation spectroscopy; molecular rotational spectroscopy; frequency locking

I. Introduction

Optical-based generation of microwave and millimeter-wave radiation has recently produced oscillators with remarkably low phase noise [1]. Such stable oscillators are potentially compatible with advanced data encoding techniques e.g., high order quadrature amplitude modulation (QAM). These oscillators utilize optical heterodyne techniques with a UTC-PD, which can generate radiation up to the THz regime [2]. Higher frequency bands may enable higher data transfer rates. The combined benefits of low phase noise and higher frequencies make optical based microwave and millimeter-wave generation key components in future communication technologies, such as 6G [3].

Future communications networks operating at higher frequencies will need standardized frequency references. Currently, there is a gap in frequency references native to this band. Either multiplication of a microwave frequency standard or division of an optical atomic clock are required to produce a suitable reference. These schemes introduce noise, cost, and complexity to potential communications system. It is therefore desirable to develop frequency references in this band that can be used to discipline oscillators. A natural choice for the task is molecular rotations. Quantized molecular rotations of small molecules in the gas phase occupy frequencies from GHz to THz and are thus excellent candidates for standardized frequency references [4].

We show the optical generation of millimeter-wave radiation together with locking to a molecular rotational transition has produced a ~300 GHz oscillator with 10^{-11} fractional frequency stability at one second averaging time.

II. Methods/Results

A schematic of the experimental setup is shown in Fig. 1. The light from two diode lasers was combined and sent on two optical paths. The first optical path interrogated the rotational absorption of N_2O molecules. The light was fed to a UTC-PD, which converted the optical power to millimeter-wave radiation, in a rough vacuum chamber (base pressure ~20 mTorr). The UTC-PD was temperature stabilized (±10 mK) by a thermoelectric cooler (TEC). The radiation passed through a low partial pressure (~50 mTorr) of N_2O (~10 cm path length) before being detected by a Schottky diode. The electrical signal from the diode was split with a bias-T into AC and DC components. The AC component contained the error signal fed back to the diode lasers through and FPGA-based instrument that performed both lock-in amplification and PID control. The DC signal drove the frequency of an RF synthesizer that could be simultaneously counted with the output of the second optical

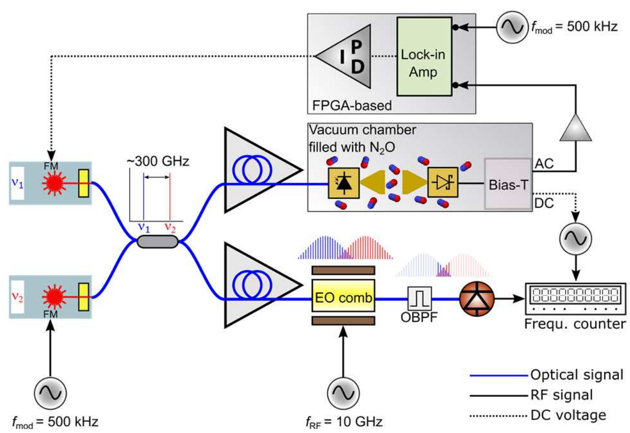

Fig. 1. Schematic layout of the experiment described in detail in the text.

path, which measured the frequency difference between the two diode lasers via EO frequency down-conversion. This was accomplished by an EO comb generated by each diode laser with ~10 GHz spacing between lines. The comb frequency spacing was set by a digital synthesizer, locked to a Rb clock reference. Overlapped high-order harmonics of each comb were isolated by an optical band pass filter (OBPF), and then photo-detected. The beat note frequency (~1 GHz) was then electronically divided and subsequently counted by a frequency counter, which was also referenced to the Rb clock.

We performed wavelength modulation spectroscopy to generate an error signal from the molecular absorption [5]. The error signal was used to lock the frequency difference between the diode lasers, and thus the millimeter-wave, to the N_2O rotational line. Once locked, the millimeter-wave oscillator frequency was counted for a few days. Fig. 2 shows the Allan deviation of the counted frequency, using an averaged one second gate time. For averaging times less than one second, a 1ms gate time was used.

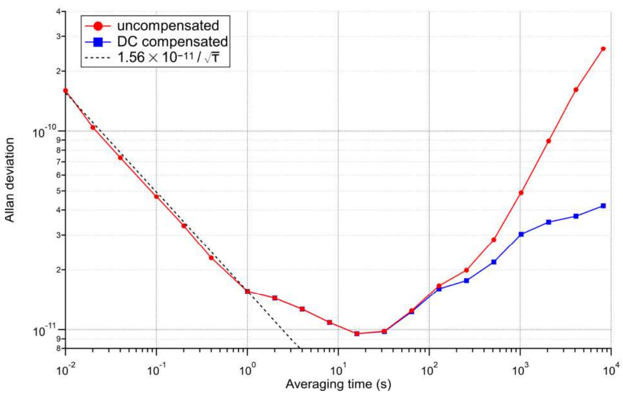

Fig. 2. Allan deviation of the millimeter wave oscillator absolute frequency. A reference line of $1.56\times10^{-11}/\sqrt{\tau}$, where τ is the averaging time, depicts the fractional stability limited by the intermodulation effect associated with the phase noise of the diode lasers. Above one second, two traces are shown. The red circles show the fractional stability of the measured frequency with no post processing. The blue squares show the improvement at long time scales after correcting frequency drifts with the DC signal from the bias-T.

Below one second averaging time, the frequency followed a slope set by the phase noise of the two diode lasers modulated at 500 kHz. Above one second averaging time, the absolute frequency drifted due to temperature fluctuations of the UTC-PD and pressure fluctuations of the N_2O gas. Both effects caused a change in the amplitude of the error signal that led to a shift in the locked frequency. They also caused a change in the DC level out of the bias T. Since the DC level was measured simultaneously with the output frequency, we could post-process the frequency data to remove long term drifts. The resulting Allan deviation of the post-processed data is shown in Fig. 2 as DC compensated. This shows almost an order of magnitude improvement at an averaging time of 10^4 s.

III. DISCUSSION/INTERPRETATION

Initial investigations into the systematics of the experiment indicate that the frequency fluctuations observed in our oscillator are not caused by the molecular rotational line itself. Rather, the fluctuations can be traced to other instabilities in our experimental setup.

Short-term frequency stability of this oscillator is dominated by the phase noise of the two diode lasers. Modulation schemes for communications experiments will likely rely on these timescales. Improvements in the frequency stability at one second averaging times and below can be achieved by using an optical setup with lower phase noise, such as a microresonator-based soliton comb [1], or by increasing the modulation frequency [5].

For applications requiring intermediate averaging times (1s-100s), the frequency fluctuations are dominated by temperature instability of the UTC-PD. Better temperature stabilization (~1mK) may be possible with the TEC. A more robust approach would include stabilization of the UTC-PD photocurrent via active feedback.

If even longer averaging times are needed, as in the case for precision molecular rotational spectroscopy, then the pressure stability of the vacuum chamber must be addressed. Here the DC level of the Schottky can be used to feed back to pressure control of the gas in the chamber. This method is superior to direct pressure measurements from a gauge because the DC level is directly sensitive to the N2O pressure in the radiation beam path. Given the constrained geometries and thus poor gas conductance of waveguide components used to work with millimeter-wave radiation, this pressure could be significantly different than the equilibrium pressure in the chamber.

IV. CONCLUSIONS

We built a millimeter-wave oscillator via optical heterodyne of two laser diodes on a UTC-PD. We then demonstrated locking of the oscillator to a molecular rotational line as a frequency reference. We measured and characterized the performance of the locked oscillator in terms of absolute fractional frequency deviations, as measured against a Rb clock. We achieved 10^{-11} stability at 1s averaging, and demonstrated a very robust lock, remaining locked and free of cycle slips for days at a time. The long-term fractional stability of the oscillator was on the order of 10^{-10} (30 Hz) or better.

The apparatus is a suitable candidate for future millimeter-wave communications studies that utilize higher frequencies and sophisticated modulation techniques. As we continue to investigate the sources of frequency drifts in our system, we aim to fully characterize the systematics of the absolute frequency of the molecular rotation. A careful analysis of the systematics and further improvements in the fractional stability could culminate in a secondary frequency standard, native to the millimeter-wave, that would be competitive with other conventionally available clocks.

REFERENCES

[1] T. Tetsumoto, T. Nagatsuma, M.E. Fermann, G. Navickaite, M. Geiselmann, and A. Rolland, "Optically referenced 300 GHz millimetre-wave oscillator," Nat. Photon., 15, pp.516–522, 2021.

[2] T. Ishibashi and H. Ito, "Uni-traveling-carrier photodiodes," J. Appl. Phys., 127, 031101, 2020.

[3] S. Dang, O. Amin, B. Shihada, and M. Alouini, "What should 6G be?," Nat. Electron., 3, pp.20–29, 2020.

[4] C.H. Townes and A.L.Schawlow, "Microwave spectroscopy," New York: Dover Publications, Inc., 1975, pp. 3-82.

[5] J. M. Supplee, E.A. Whitaker, and W. Lenth, "Theoretical description of frequency modulation and wavelength spectroscopy," Appl. Opt., 33, pp. 6294-6302, 1994