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IN THE 225-315 GHz RANGE***

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Open-Path Millimeter-Wave Spectroscopy in the 225-315 GHz Range

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ABSTRACT

This paper discusses the development of an open-path millimeter-wave (mm-wave) spectroscopy system in the 225-315 GHz atmospheric window. The new system is primarily a monostatic swept-frequency radar consisting of a mm-wave sweeper, hot-electron-bolometer or Schottky detector, and trihedral reflector. The heart of the system is a Russian backward-wave oscillator (BWO) tube that is tunable over 225-350 GHz. A mm-wave sweeper has been built with the BWO tube to sweep the entire frequency range within 1 s. The chemical plume to be detected is situated between the transmitter/receiver and the reflector. Millimeter-wave absorption spectra of chemicals in the plume are determined by measuring swept-frequency radar signals with and without the plume in the beam path. Because of power supply noise and thermal instabilities within the BWO structure over time, the BWO frequencies fluctuate between sweeps and thus cause errors in baseline subtraction. To reduce this frequency-jitter problem, a quasi-optical Fabry-Perot cavity is used in conjunction with the radar for on-line calibration of sweep traces, allowing excellent baseline subtraction and signal averaging. Initial results of the new system are given for open-path detection of chemicals.

Key Words: Millimeter waves, backward-wave oscillator, molecular spectroscopy, open-path monitoring.

1. INTRODUCTION

Under the 1990 Clean Air Act Amendments, industry is required to demonstrate environmental compliance by enhanced (continuous) monitoring of its emissions or of parameters correlated to its emissions.¹ The continuous monitoring devices (CEMs) available to date are mostly species-specific and use sampling techniques. Standoff or open-path monitoring of stack emissions is attractive because it is nonintrusive and obviates the need for sampling. Electromagnetic radiation utilizing broadband sources in the infrared (IR), visible, and ultraviolet (UV) regions has been investigated for remote sensing of chemicals.² In addition, laser techniques known as LIDAR (light detection and ranging) are gaining importance for remote sensing.³ These techniques are generally based on molecular vibrational energy transitions in the IR region, electronic energy transitions in the UV and visible regions, and Raman scattering in the UV region.

Millimeter-wave (mm-wave) spectroscopy based on rotational energy transitions is a complementary technique to the optical and laser approaches. Because of their longer wavelengths compared to those optics, mm-wave techniques can be used in smoky and dusty environments, are less affected by cloud conditions, and can provide longer detection ranges. Additionally, interference from water vapor and other common atmospheric species such as CO₂ and methane is reduced when using mm

waves. Polar molecules selectively absorb electromagnetic radiation of specific wavelengths in the centimeter and millimeter regions in accordance with their rotational energy transitions.⁴ Millimeter-wave spectral lines are narrow and highly resolvable under low pressures (<1 torr). As the pressure increases, however, the spectral lines are broadened due to molecular collisions, and also the peaks of the lines are reduced. As a result, detection sensitivity and chemical selectivity suffer at higher pressures. The pressure-broadening problems inherent with open-path monitoring (e.g., in the atmosphere) can be mitigated by using high-frequency mm-wave sources with wide sweep-width capability. Detection sensitivity increases in general with the square of the frequency, and the number of resolution lines (chemical selectivity) increases with the sweep width.

Recently, feasibility of the mm-wave technique was shown for environmental monitoring in open air by using a monostatic swept-frequency radar system in the 225-315 GHz range.⁵ The mm waves were generated by employing a low-frequency sweeper and frequency triplers. Because the triplers, which are based on harmonic generation are not efficient, the radiation power obtained was low in the microwatt range. The narrow bandwidth of the triplers also limited the tuning range to within 50 GHz. The proof-of-principle was tested with this system by detecting D₂O in open air. In addition, the chemical selectivity of the technique was demonstrated by employing a special deconvolution procedure; individual chemicals in a multicomponent mixture were determined with both simulated and experimental spectra.⁶

This paper describes the development of a prototype radar system based on a Russian backward-wave-oscillator (BWO) tube (Model OB-30) that can sweep the entire frequency range within 1 s and generates up to 10 mW of power. This higher power allows longer detection range and the wider tuning range improves spectral line resolution and molecular selectivity. Because of power supply noise and thermal instabilities in the BWO structure over time, the BWO frequencies fluctuate between sweeps and cause poor baseline subtraction. To mitigate the frequency jitter problem, part of the mm-wave beam is coupled to a quasi-optical Fabry-Perot cavity that produces a sequence of frequency markers during the sweep. These cavity markers are used in software to produce on-the-fly calibration of the sweep traces, which then allows excellent baseline subtraction and signal averaging. Initial results of the new mm-wave system are given for open-path detection of chemicals.

2. DEVELOPMENT OF MONOSTATIC RADAR SYSTEM

Figure 1 is a schematic diagram of the monostatic radar system built for open-path chemical measurements in the frequency range of 225-315 GHz. It consists of a free-running mm-wave sweeper that generates and transmits mm waves through a plume of chemicals to be measured. A retroreflector is used on the far side of the plume to return the radiation to the transmitter. A liquid-helium-cooled bolometer or ambient-temperature Schottky detector is used to measure return signals. Absorption spectra of the plume gases are determined by comparing the return signals with and without the plume in the beam path.

2.1 Millimeter-wave sweeper

A backward-wave oscillator tube (OB-30 from the Istok Company, Russia) is the heart of mm-wave sweeper. The tube, as shown in Fig. 2(a), is 25 mm in diameter and 35 mm long, and water cooled. It requires an AC filament power supply (6.4 V, 1.35 A) to heat the cathode, a high-voltage (HV) power supply (1000-4000 V, 40 mA) between the cathode and slow-wave structure to accelerate the electrons, and a DC magnetic field of 7000 G between cathode and collector to steer the electron beam over the slow-wave structure. A permanent magnet made of cobalt-samarium alloy produces the magnetic field. To generate mm-wave oscillations, the tube must be properly aligned with respect to the DC magnetic field and is mounted in the pole gap of the magnet with a three-axis alignment fixture, see Fig. 2(b); the radiation is coupled to a scalar horn through an over-moded rectangular wave guide attached to the tube structure. The tube is tunable from 225 to 350 GHz by varying the HV from 1000 to 4000 V and provides average of 10 mW. A 5-kV Bertan power supply (Model 210-05R) that can be swept by a 0-5 V remote control input is used to sweep the HV.

Fig. 1. Schematic diagram of mm-wave radar system in 225-315 GHz range.

The BWO tube generates monochromatic radiation, but its frequency stability depends on (a) power supply voltage fluctuations, (b) alignment of the tube in the magnetic field, and (c) temperature of the tube structure. The frequency of the tube is controlled mainly by the HV power supply. On the average, the tube has a sensitivity of 70 MHz/V. Because the BWO tube has extremely low internal capacitance, all HV noise, including 60-Hz ripple and switching power-supply noise (20 kHz), are converted into frequency variation. Three types of instabilities (blinking effect, mode breaks, and mode switching) may occur while tuning the tube, but in most cases can be eliminated by proper alignment. To control the temperature of the tube structure and thus ensure long-term stability of the radiation, a temperature-controlled closed-cycle water pump circulates coolant through the tube.

(a) Backward-wave oscillator (BWO) tube

(b) BWO tube positioned inside magnet, with alignment fixture

Fig. 2. Backward wave oscillator: (a) internal structure of tube, (b) support and alignment fixture inside the permanent magnet.

Despite of all the precautions one may take to control BWO stability, it is nearly impossible to achieve the frequency stability required for baseline subtraction and signal averaging. Hence, one often resorts to phase-locked control of the BWO in high-resolution spectroscopy, but this is rather slow for

cavity markers. Because the cavity markers correspond to the actual frequencies during a sweep, the cavity markers and the corresponding samples of radar data are aligned or matched with each other by software between data traces. The radar data between the cavity markers are interpolated linearly. Figure 4 shows subtraction of two BWO swept-frequency traces, with and without the use of cavity markers. Because of transients associated with the return ramp of the sweep, the first 5000 points of 25,000 points of the sweep trace are ignored. Further improvement is possible by increasing the cavity length, which in turn decreases the spacing between frequency markers. Thus, the use of an F-P cavity enables us to calibrate on-the-fly the frequencies of a free-running oscillator, providing excellent baseline subtraction and signal averaging.

Fig. 4. Baseline subtraction with and without cavity markers.

To test the spectral line detection, 200 mtorr of CH_3CN was pumped into a 1.4-m-long gas cell. The BWO was swept linearly between 1500 to 2700 V in 1 s. Three groups of CH_3CN lines around 276, 294, and 312 GHz were covered in the sweep range. Figure 5 gives the percent absorption of the gas determined by measuring the detected power (with a bolometer) with and without the gas in the cell. The small spikes along the baseline correspond to those cavity lines not totally subtracted out. The sweep trace was calibrated in terms of frequency by using the known CH_3CN lines and appropriate interpolation based on the BWO voltage-to-frequency static tuning curve.

Resolution and sensitivity of the system can be determined by examining (zooming) a narrow frequency range of the fast sweep data. Figure 6 is a replot of the data in Fig. 5 over a 5 GHz range near the 276 GHz line. We see not only the rotational lines of the ground state but also those of excited vibrational states. The system provides a resolution of about 4 MHz and a sensitivity of 10^{-3} cm^{-1} in the fast-sweep mode for low-pressure lines.

Fig. 5. Low-pressure spectra of CH_3CN .

3. Open-path measurements

The monostatic mm-wave radar system in Fig. 1 was tested for open-path detection of chemicals in the laboratory, with a distance of 5 m between the transmitter and reflector. The same sweep parameters (sweep range and sweep rate) used for the CH_3CN tests (Fig. 5) were adopted for this test, so that the calibration of the sweep trace is known with respect to frequency. A plume was created by heating D_2O in a beaker and releasing the vapor in the path of the mm-wave beam. To improve the pathlength, we contained the plume in a 46-cm-long T-shaped glass pipe with open ends. Figure 7 shows the absorption peak of D_2O around 318 GHz. Fluctuation in the baseline is due to changes in the pattern of the standing waves that arise from reflections at the detector, reflector, and transmitter.

Fig. 6. Replot of Fig. 5 over a narrow frequency range.

4. CONCLUSIONS

We have developed a swept-frequency monostatic radar system in the range of 225-315 GHz for open-path detection of chemicals. As a part of the system, a mm-wave sweeper has been built using a Russian backward-wave oscillator (BWO) tube; it can be swept over the entire frequency range in 1 s and provides an average power of 10 mW. While a free-running BWO sweeper is inherently unstable in terms of repeatability of frequencies during a sweep, this frequency instability problem was solved by using a Fabry-Perot cavity that provides on-the-fly calibration of frequencies. The BWO sweeper was tested by measuring low-pressure CH_3CN lines in a gas cell. A comparison of baseline subtraction with and without the cavity markers showed the superiority of the cavity approach. The system showed a frequency resolution of 4 MHz and a detection sensitivity of 10^{-3} cm^{-1} in a fast scan of low-pressure lines. The new system offers a major improvement in power, sweep range, and detection sensitivity over the previous system which used frequency triplers on the output of a low-frequency sweeper.⁵

Open-path detection capability of the monostatic swept-frequency radar system was tested by releasing D_2O in open air. Initially, the system was tested in the laboratory over a short range (5-m distance between the transmitter and reflector). The tests showed the detection of D_2O absorption line

at 318 GHz. Further work is underway to test the system at longer distances and against different chemicals by sampling chemicals into a plastic (Tedlar) bag and placing it in the beam path.

Fig. 7. Detection of D₂O plume released in open air.

5. ACKNOWLEDGMENTS

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