Tunable far-infrared spectroscopy

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Tunable, cw, far-infrared radiation has been generated by nonlinear mixing of radiation from two CO_2 lasers in a metal-insulator-metal (MIM) diode. The FIR difference-frequency power radiated from the MIM diode antenna to a calibrated indium antimonide bolometer. Two-tenths of a microwatt of FIR power was generated by 250 mW from each of the CO_2 lasers. The combination of lines from a waveguide CO_2 laser, with its larger tuning range, with lines from CO_2, N_2O, and CO_2 isotope lasers promises complete coverage of the entire far-infrared band from 100 to 5000 GHz (3–200 cm^{-1}) with stepwise-tunable cw radiation. To demonstrate the usefulness of the technique, we observed the $J = 4$–5 line of CO at 567 GHz.

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Lasers are providing cw tunable radiation in most of the electromagnetic spectrum; however, tunable spectroscopy in the far-infrared (FIR) region has only recently been accomplished by the addition of microwave sidebands to radiation from a FIR laser,[1,2] by harmonics of klystrons and backward wave oscillators,[3,4] by backward wave oscillators themselves,[5] and by CO_2 difference-frequency generation in GaAs.[6] We report the generation of tunable cw FIR radiation (TuFIR) via CO_2 difference frequency mixing in a W-Ni metal-insulator-metal (MIM) diode. This discovery promises complete coverage of the FIR frequency region with the additional advantages of simplicity and high accuracy.

Previously, the MIM diode had been used mainly to make direct frequency measurements of frequencies from 0.3 to 200 THz (i.e., wavelengths from 1000 to 1.5 μm). The MIM diode also had been used to add microwave sidebands onto CO_2 laser radiation.[7] The power of the propagating sideband radiation at 10 μm was small (5×10^{-11} W), probably because the antenna properties of the diode were not well known at that time and coupling to the diode was weak. Only recently have significant improvements in coupling and stability[8] encouraged attempts to use the diode for the generation of FIR radiation.

The MIM diode was first reported in 1966,[9] and its use for laser frequency measurements with pulsed lasers was reported in 1969.[10] Subsequently, it was found to be very useful for frequency measurements of cw radiation at frequencies up to 200 THz.[11–13] The diode has also been used to make frequency-difference measurements in the visible.[14] Tunneling is apparently responsible for the operation of this extremely useful diode[15–17]; however, definitive proof of the mechanism has not yet been made.

Two conventional sealed-off CO_2 lasers with grating line selectors and PZT tuners provided the cw radiation for the initial experiment. A power of 250 mW from each laser was focused colinearly on a conventional W-NiO-Ni MIM diode. The CO_2 radiation was focused on the conical tip of

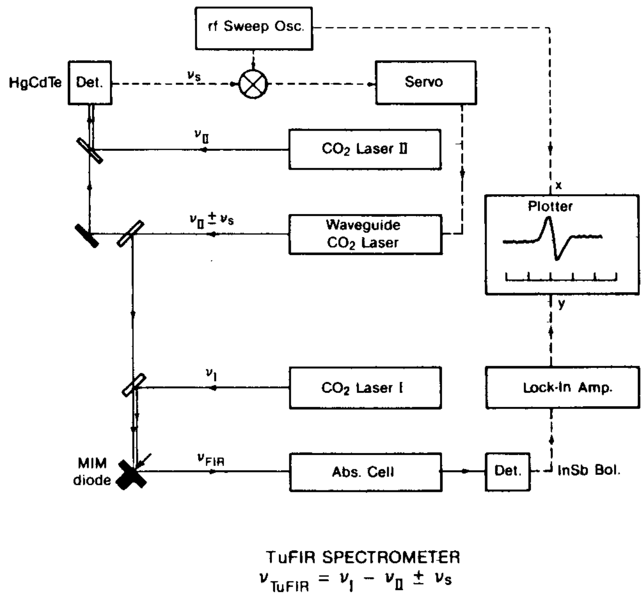

FIG. 1. Block Diagram of TuFIR spectrometer.

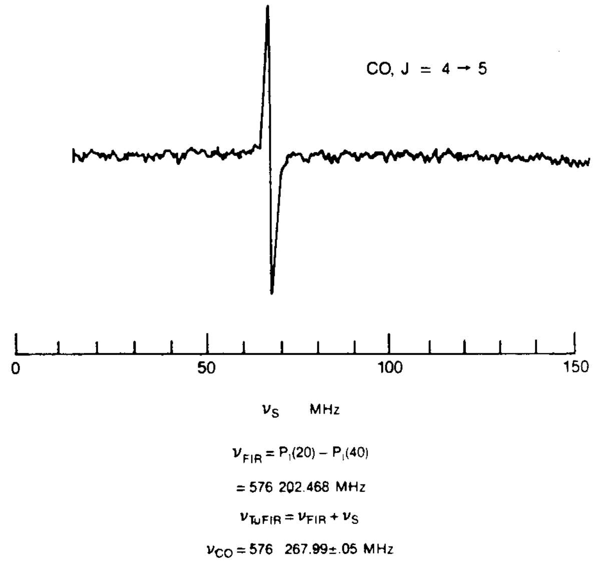

FIG. 2. CO absorption derivative signal for the $J = 4$–5 transition of CO. Data were taken with a CO pressure of 5.3 Pa and a time constant of 400 ms.

the tungsten antenna and produced 20 mV of rectified signal with a diode impedance of 120 Ω. The lasers were set to CO_2 lines separated by 570 GHz.

The entire 10-mm length of the 25-μm-diam tungsten wire radiated 570-GHz power from the diode in a conventional long-wire-antenna pattern. This radiation was collimated with a 12.7-mm-diam circular, 45° segment of an off-axis 9-mm focal length parabola located 18 mm from the center of the antenna. This arrangement effectively collimated the radiation, which propagated 5 cm to the mouth of the 9-mm-diam conical horn of the helium-cooled InSb bolometer. A maximum of 0.2 μW of TuFIR was detected. Quartz and polyethylene filters were inserted in the beam to verify that the frequency was approximately at 0.5 THz ($\lambda = 0.6$ mm).

The InSb bolometer was calibrated with FIR radiation from a 570-μm methanol laser pumped with the $P_{II}(16)$ line of a CO_2 laser. A 12.7-mm-thick calibrated attenuator of acrylic resin was used to decrease the FIR power from 1.5 mW to prevent saturation of the InSb detector. The unattenuated power was measured in an electrically calibrated cone calorimeter.[18] From this measurement, the NEP of the InSb detector was found to be 1.4×10^{-11} W $\pm$ 20% at 570 μm for a 1-s time constant. This number is in fair agreement with that provided by the manufacturers.

To see if radiation could be generated at higher frequencies, we selected CO_2 lines with frequency separations increasing up to 3.5 THz (80 μm). A He-cooled Ge bolometer with a shorter wavelength response was substituted for the InSb detector for frequencies above one terahertz. Far-infrared radiation was observed from all oscillating pairs of CO_2 laser lines, indicating that FIR radiation generation is possible at least to 3.5 THz.

As further proof that indeed we have FIR radiation and to demonstrate the usefulness of this new technique, we observed the $J = 4$–5 transition of CO in absorption. Figure 1 shows the experimental setup. Laser I was tuned to $P_I(40)$ and laser II to $P_I(20)$ in $^{12}C\,^{16}O_2$. Both lasers were stabilized to saturated-absorption lines in a low-pressure CO_2 cell. A waveguide CO_2 laser tuned to $P_I(20)$ was frequency-offset locked to laser II via a voltage-sweepable rf oscillator. Beams from the waveguide laser and laser I were combined and focused on the MIM diode. The waveguide laser could be tuned more than $\pm$ 120 MHz, and hence, the FIR frequency could be swept by that same amount. The center frequency ν_{FIR} is the difference between the frequencies of the two locked CO_2 lasers:

$$\nu_{FIR} = P_I(20) - P_I(40) = 576\,202.468 \text{ MHz}.$$

Then, the TuFIR frequency, ν_{TuFIR}, is given by

$$\nu_{TuFIR} = \nu_{FIR} \pm \nu_s ,$$

where ν_s is the beat frequency between the line-center-locked and waveguide lasers. A 1-m copper tube, 14 mm in diameter, with polyethlene windows, was used as a waveguide absorption cell and was placed between the MIM diode and the InSb detector. A typical trace is shown in Fig. 2. The frequency at the zero crossing of the derivative is 576 267.99 $\pm$ 0.05 MHz, in good agreement with previous work.[19]

In previous frequency-measuring experiments in our laboratory, we have used the MIM diode to synthesize harmonic currents at five times the frequency of a CO_2 laser, indicating that the response frequency of the diode is greater than 150 THz (i.e., the response time $< 0.7 \times 10^{-14}$ s). Thus, we think that sum-frequency radiation is probably also present in significant quantities. The detection of the sum radiation, however, requires different techniques because of the large amount of room-temperature blackbody background radiation at these wavelengths.

In conclusion, we have generated spectroscopically significant amounts of tunable far-infrared radiation using low-power CO_2 lasers and a simple room-temperature point-contact diode. This discovery should greatly facilitate far-infrared spectroscopy.

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