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Coherent Tunable Far Infrared Radiation*

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Abstract. Tunable, cw, far infrared (FIR) 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 was 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. Using 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.

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Lasers provide cw tunable radiation over some parts 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 byCO_2 difference-frequency generation in GaAs [6]. We describe the generation of tunable cw FIR radiation (TuFIR) by means of 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 (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 as large as 2.5 THz in the visible [14]. Tunneling is apparently responsible for the operation of this extremely useful diode [15–17]; however, definitive proof that this is the mechanism has not yet been found.

Experimental

Two FIR laser spectrometers are now in operation at the National Bureau of Standards, Boulder Laboratories. The FIR radiation is synthesized in two ways: either from the difference between the CO_2 laser frequencies (second order), or, from the difference between two CO_2 lasers with the addition of microwave sidebands (third order).

In the first method

$$\nu_{\mathrm{FIR}} = \nu_I - \nu_{II}, \tag{1}$$

where ν_I us the frequency of a stabilized, single-mode CO_2 laser, and ν_{II} is the frequency of a tunable CO_2 waveguide laser which is referred to a third stabilized single-mode CO_2 laser for its frequency control. This method was first described by Peterson et al. [18] and

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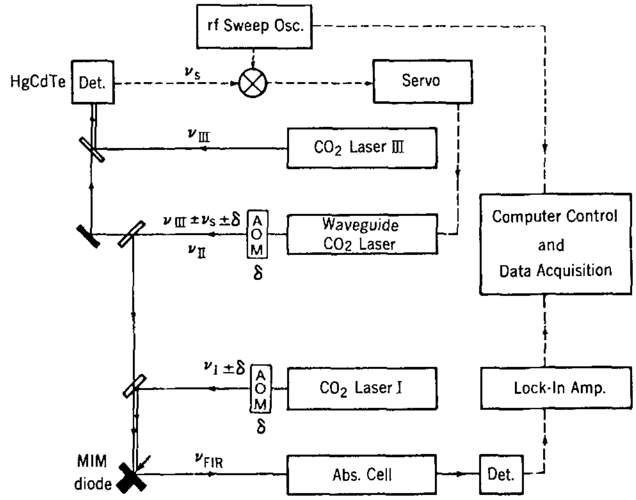

Fig. 1. Tunable FIR spectrometer which uses a waveguide laser for the tunable frequency component. This system corresponds to two-wave or second-order mixing. The acousto-optic modulators (AOM) shift the laser frequency by $\pm\delta$, and reduce the level of feedback-excited power fluctuations of the CO_2 laser. The frequency ν_s is an rf offset frequency

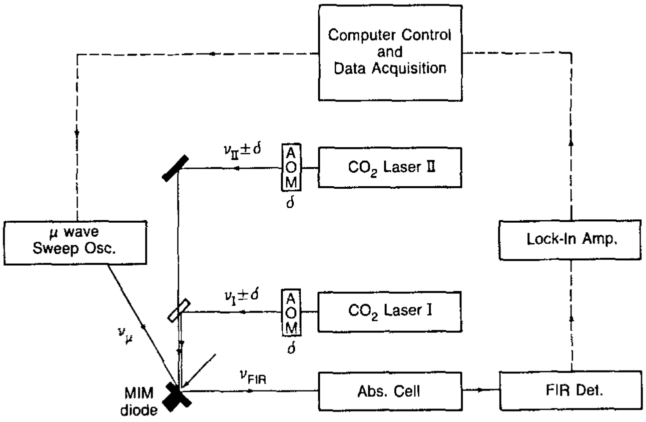

Fig. 2. Tunable FIR spectrometer which uses an rf frequency synthesizer for the tunable frequency component. This system corresponds to three-wave or third-order mixing

is illustrated schematically in Fig. 1. Here the mixing is second order (two wave) in a metal-insulator-metal (MIM) diode, and the FIR frequency is the frequency difference between the two CO_2 lasers which are incident on the MIM diode.

Our second method employed third-order (three wave) mixing, and the relevant schematic diagram is shown in Fig. 2. Here two fixed-frequency, stabilized CO_2 lasers, operating at frequencies ν_I and ν_{II}, and a tunable microwave component at frequency ν_μ all have their radiation coupled to a MIM diode. The free frequencies are combined to produce a FIR frequency in this case given by

$$\nu_{FIR} = (\nu_I - \nu_{II}) \pm \nu_\mu \,. \qquad (2)$$

A microwave sweeper provides up to ± 20 GHz tunability, where one of the microwave sidebands is selected to sweep the spectral region of interest. However, the advantage of broad tunability is somewhat offset because third-order mixing has lower FIR power than second-order mixing.

In both systems the metal-insulator-metal diode is a 25 μm diameter, pointed tungsten wire contacting the polished end of a metal substrate at normal incidence. The properties of this contact with the metal post provide the nonlinear current-voltage characteristic which generates the FIR radiation. The tungsten wire which is several millimeters in length functions as a long wire antenna for the FIR radiation. The FIR radiated power is reflected by a section of a parabolic reflector and propagated along the axis of a single-pass Pyrex absorption cell (10 to 50 cm long and 19 mm inside diameter). The cell has high density polyethylene windows, and the transmitted power is sensed by an appropriate detector. The amount of FIR power at the detector depends upon several variables. To prevent diode damage, the total CO_2 power at the MIM diode is generally limited to about 200 mW; in second order, this results in 100 to 500 nW of FIR radiation. In third order, the FIR power is typically one-third of this amount.

The optimization of the diode junction remains poorly understood and is largely a matter of experimental practice. Our best junctions for second-order operation use nickel bases. For third-order mixing we have less experience, but contacts to cobalt substrates are better than those to nickel. Considerable patience is required, especially in third order, as the whisker contact needs careful and repeated adjustment to maximize the FIR power.

The FIR radiation is frequency modulated at a 1-kHz rate with a typical amplitude of 2.5 MHz by application of an ac voltage to the piezoelectric-transducer-frequency-stabilization control of one of the CO_2 lasers. The modulated FIR frequency is scanned across the spectral region of interest by one of the two techniques described previously, and the resulting FIR signal is the modulation-broadened first derivative of the absorption as a function of frequency. The center zero signal crossing position indicates the absorption line-center frequency and the absolute maxima of this signal correspond to the inflection positions of the absorption profile. A measured spectrum using third-order mixing is shown in Fig. 3.

The signal-to-noise ratio of the absorption profile depends upon several factors. The FIR power varies with each contact of the diode, and the noise due to the FIR power fluctuations is often greater than the intrinsic detector noise, although some recent refinements have improved the overall performance. First,

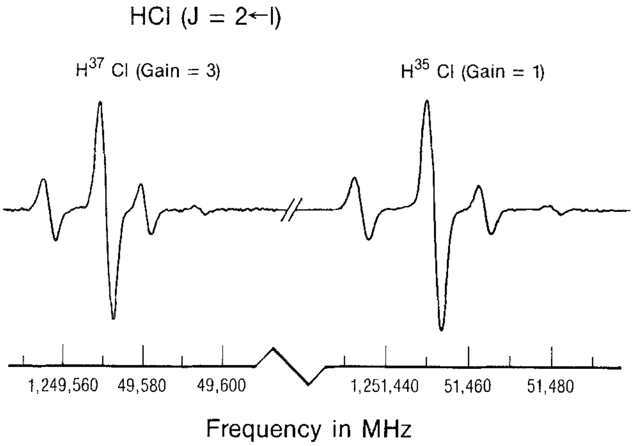

Fig. 3. Measurement of the $J=2\leftarrow1$ transition of HCl illustrating the wide scanning capability and the high accuracy of the third-order spectrometer. In this case, eight HFS transitions for each isotope are seen as four features. For this measurement we used a pumped, ^{4}He-cooled germanium bolometer and preamp system which was engineered for optimum response at the signal modulation of 1 kHz. The scan for each isotope is an average of two frequency scans over the indicated ranges in a total time of 3.5 min

acousto-optic couplers have been added to isolate the CO_2 lasers from the diode. This isolation serves to reduce power fluctuations caused by feedback in the CO_2 laser. Second, detector systems have been optimized for phase-sensitive detection of the FIR at a modulation frequency of 1 kHz, which is a reasonable trade-off between $1/f$ noise and detector responsivity. With the best second-order diodes, the laser-driven FIR intensity fluctuations exceed the detector noise; however, in third order, overall noise is dominated by detector noise because the FIR power is less. In addition to the detector noise on the signal, the crossing point can also be perturbed by transient power changes and standing wave effects between the diode and the detector. The latter effects are suppressed in part by mounting all optical transmission elements (windows, filters, detectors) at small angles from normal incidence to the radiation direction. A typical signal-to-noise ratio with a 1 Hz bandwidth is on the order of 10^3 to 10^4 for a 10% absorption.

The uncertainty in the measurement of a line center position is 0.1 to 0.2 MHz under conditions which give good signal-to-noise ratios. This uncertainty must be combined with the uncertainty of the FIR frequency, which is the combined quadratic sum (typically 35 kHz) of the uncertainties of the two CO_2 laser line frequencies. The 35 kHz uncertainty reflects the realization of CO_2 frequencies [19] using our saturated absorption stabilized lasers. Only in the case of the strongest, narrowest lines was the line center measured with sufficient accuracy that the underlying uncertainty of the FIR frequency affected the accuracy of the final measurement.

Conclusions

Further improvements in this difference technique for the generation of tunable FIR radiation may come from either improved diodes or detection schemes. The non-linearities measured in the current-voltage curves of our MIM diodes are extremely small and conversion efficiencies could be 100 times larger. We are optimistic that better materials which will result in larger FIR powers may be found. Differential detection schemes would also significantly improve the sensitivity and permit the detection of weaker lines.

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References

1. D.D. Bicanic, B.F.J. Zuidberg, A. Dynamus: Appl. Phys. Lett. **32**, 367 (1978)
2. H.R. Fetterman, P.E. Tannenwald, B.J. Clifton, W.D. Fitzgerald, N.R. Erickson: Appl. Phys. Lett. **33**, 151 (1978)
3. P. Helminger, J.K. Messer, F.C. DeLucia: Appl. Phys. Lett. **42**, 309 (1983)
4. E.A. Cohen, H.M. Pickett: Molec. Spectrosc. **93**, 83 (1982)
5. A.F. Krupnow: In *Modern Aspects of Microwave Spectroscopy*, ed. by G.W. Chantry (Academic, New York 1979) p. 217
6. R.L. Aggarwal, B. Lax, H.R. Fetterman, P.E. Tannenwald, B.J. Clifton: J. Appl. Phys. **45**, 3972 (1974)
7. A. Sanchez, S.K. Singh, A. Javan: Appl. Phys. Lett. **21**, 240 (1972)
8. K.M. Evenson, M. Inguscio, D.A. Jennings, F.R. Petersen: J. Appl. Phys. **57**, 956 (1985)
9. J.W. Dees: Microwave J. **9**, 48 (1966)
10. V. Daneu, D. Sokoloff, A. Sanchez, A. Javan: Appl. Phys. Lett. **15**, 398 (1969)
11. K.M. Evenson, J.S. Wells, L.M. Matarrese, L.B. Elwell: Appl. Phys. Lett. **16**, 159 (1970)
12. K.M. Evenson, J.S. Wells, F.R. Petersen, B.L. Danielson, G.W. Day: Appl. Phys. Lett. **22**, 192 (1973)
13. D.A. Jennings, F.R. Petersen, K.M. Evenson: In *Laser Spectroscopy IV*, ed. by H. Walther, K.W. Rothe (Springer, Berlin, Heidelberg 1979) p. 39
14. R.E. Drullinger, K.M. Evenson, D.A. Jennings, F.R. Petersen, J.C. Bergquist, L. Burkins: Appl. Phys. Lett. **42**, 137 (1983)
15. S.M. Faris, T.K. Gustafson, J.C. Wiesner: IEEE J. QE-**9**, 737 (1973)
16. G.M. Elchinger, A. Sanchez, C.F. Davis, Jr., A. Javan: J. Appl. Phys. **47**, 591 (1976)
17. A. Sanchez, C.F. Davis, K.C. Liu, A. Javan: J. Appl. Phys. **49**, 5270 (1978)
18. K.M. Evenson, D.A. Jennings, F.R. Peterson: J. Appl. Phys. **44**, 576–578 (1984)
19. F.R. Peterson, E.C. Beaty, C.R. Pollock: J. Molec. Spectrosc. **102**, 112–122 (1983)

L.C. Bradley, K.L. Soohoo, C. Freed: IEEE J. QE-**22**, 234–267 (1986)