Generation of tunable laser sidebands in the far-infrared region

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Continuously tunable laser sidebands have been generated by mixing radiation from an optically pumped far infrared (FIR) molecular laser, operated at 693, 762, 1627, and 1839 GHz, with that from millimeter-wave klystrons in a Schottky-barrier diode. An enhancement in conversion efficiency over similar systems reported previously is obtained by using a Michelson interferometer to separate the sidebands from the carrier and by placing the Schottky diode in an open structure corner cube mount. With 4 mW of laser power at 693 and 762 GHz the sideband power was measured to be 3.0 μW. This is at least an order of magnitude better than the previously reported results. At higher frequencies, 22 mW of 1627-GHz laser power produced about 2.5 μW of sideband output, while 3mW of 1839-GHz laser power generated about 100 nW of sideband radiation. The lower efficiency at the higher frequencies is due primarily to the mismatch between the laser radiation and the fixed-length diode antenna. To demonstrate the tunability of the generated far-infrared radiation, the laser sidebands were swept through absorption lines of HDO and H_2CO near 600 and 800 GHz. The absorption signals were easily seen, using either video or lock-in detection techniques.

In recent years several techniques have been developed which generate tunable coherent radiation at submillimeter and far-infrared (FIR) wavelengths. For example, harmonic generation of conventional microwave sources has produced spectrometers capable of continuous operation to above 1000 GHz, but the sensitivity of such instruments drops rapidly with frequency.[1,2] Laser-based methods, which could cover the entire FIR region, have therefore received a great deal of attention.

Nonlinear mixing techniques such as difference frequency generation from high-power CO_2 lasers in GaAs,[3] parametric oscillation,[4] and stimulated Raman scattering[5] have all produced small amounts of tunable FIR radiation. More recently, Evenson *et al.*[6] have generated spectroscopically useful amounts of tunable FIR radiation ($\sim$200 nW) by mixing two low-power CO_2 lasers in a metal-insulator-metal (MIM) diode. The limited tuning range of the individual infrared transitions means that a large number of laser lines, including isotopic species, must be available to provide complete coverage in the FIR. Alternatively, similar amounts of tunable FIR radiation ($\sim$100 nW) have been produced by mixing FIR molecular lasers and conventional microwave sources in both open[7] and closed[8] mixer mounts. We report here improvements in this approach which yield approximately thirty times more output power than previous results.

Figure 1 shows the optical layout of our laser sideband system. A commercial CO_2 pumped far-infrared molecular laser (Apollo FIR laser system, model number 560) is used as the fixed frequency source. The CO_2 laser is stabilized using an external Fabry–Perot etalon.

The FIR laser output passes through a polarizing Michelson interferometer, similar in design to that originally proposed by Martin and Puplett.[9] The analyzer polarizer and the path differences are set to pass the laser radiation without attenuation or polarization change. The polarizers are polyester-backed wire grids and exhibit a 10% loss. An off-axis parabolic mirror focuses the output radiation onto the mixer. The laser radiation is quasi-optically collected and coupled onto the mixer by a 1.7-mm-long phosphor bronze whisker antenna ($\sim 4\lambda$ at about 700 GHz), which contacts a GaAs Schottky-barrier diode located at the apex of an open structure corner cube similar to that originally proposed by Sauter and Schultz.[10,11] The diode (Mattauch No. 1V1, 5-fF capacitance, 7-Ω spreading resistance) is soldered onto a post which passes through the center of the corner cube's V-band waveguide, thereby coupling millimeter-wave radiation into the diode to be mixed with the laser. The millimeter-wave radiation is produced by a klystron phase locked to a stabilized X-band source. A ferrite modulator chopped the klystron power reaching the diode at a frequency best suited to the detector used to monitor the sidebands.

The unused laser power and the sideband power returns along the same optical path to the interferometer. The interferometer path difference is tuned so that the sideband polarization is rotated by 90° as it passes through the interferometer while the laser polarization is unrotated. This polarization behavior is accomplished when

$$2l = 2n\lambda_{\text{laser}} \cong (2m+1)\lambda_{\text{sideband}},$$

where l is the path difference between the two arms of the interferometer, and m and n are integers. Since the sideband wavelength is not necessarily harmonically related to the laser wavelength, the pathlength difference is tuned to optimize performance. The tuning is not critical since the transmittance of the Michelson interferometer is sinousoidal with l.

When polarization of the laser is 90° with respect to the diode antenna, the relation for l can be modified so that the first pass through the interferometer produces a 90° polarization change. On the return pass, the laser power undergoes a

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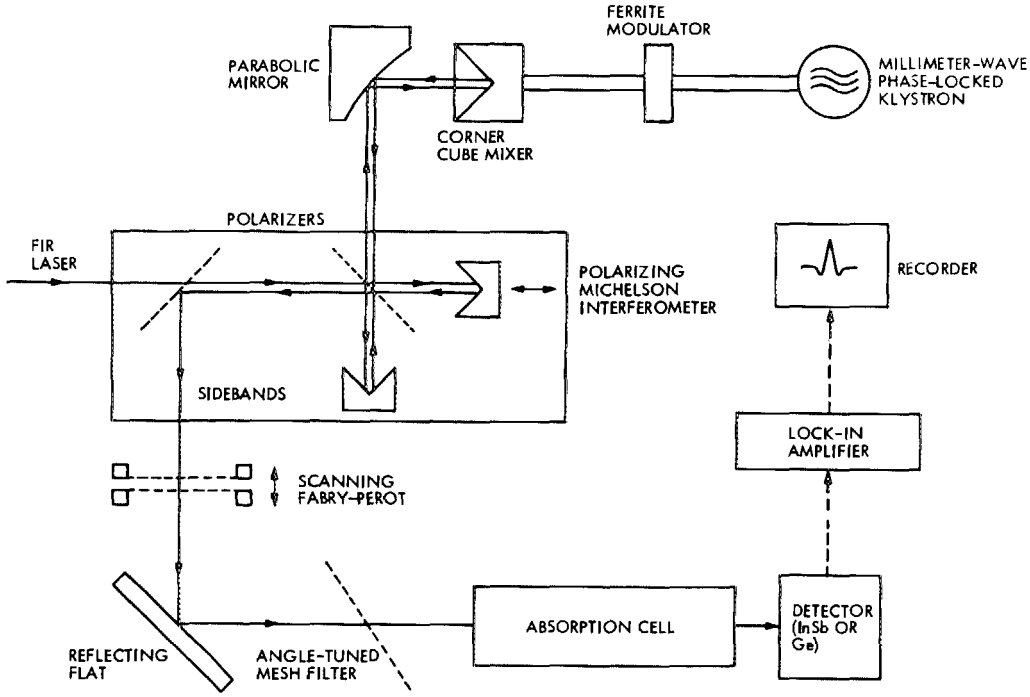

FIG. 1. Optical layout of the FIR laser sideband system.

reciprocal polarization shift, while the polarization of the sidebands remains unchanged. Thus, the optical path of the laser and sidebands is the same, regardless of the laser polarization.

In an ideal Michelson interferometer, the sideband power would all be reflected through the output port. However, because of the imperfect polarizers and possible misalignment of the interferometer, some laser power leaks through along with the sidebands. To isolate the sideband radiation from this residual laser power, the output radiation is sent through a Fabry-Perot interferometer with a finesse of about 15. When scanned, the Fabry-Perot interferometer separates the laser, as well as the upper and lower sidebands, from each other. In order to further reduce the laser power level reaching the detector, an angle tunable free-standing mesh filter is employed. Such a filter is, essentially, a very sharp notch (reject) filter whose rejection frequency is tunable by means of varying the radiation angle of incidence.[12,13]

Initially, experimental efforts were devoted to the generation and characterization of the laser sidebands as a function of several variables of the system. For this purpose, the FIR laser was operated at the 693-GHz formic acid line as the center frequency, and a 93-GHz klystron provided the tunable millimeter-wave source. The klystron power was modulated at 1 kHz, and an InSb hot-electron bolometer was placed after the tunable mesh filter with its output sent to a lock-in amplifier. Linear sweeps of the Fabry–Perot interferometer clearly indicated the generation of both sidebands with signal-to-noise ratio of better than 100. With maximum klystron power, only 50 mV of constant voltage diode bias was needed to optimize the sideband signal. Lower klystron power settings required higher external biasing for optimum sideband generation, as expected. The effect of klystron power on the sideband output was studied by monitoring the klystron power level with a second diode (Baytron model No. 1N-58). The result is the saturation curve shown in Fig. 2. The dependence of the sideband power on the incident laser power was also determined by attenuating the laser radiation with calibrated mesh filters, resulting in the linear curve also shown in Fig. 2. These results are all consistent with those reported by Bicanic.[14]

In order to determine the absolute sideband power generated, the InSb detector was calibrated against a Scientech thermopile power meter (model 361) at four different laser frequencies: 604, 693, 762, and 1367 GHz. Foote *et al.*[15] reported that the Scientech power meter response at 671 GHz is 0.52 ± 0.25 and at 1363 GHz is 0.575 ± 0.125 of the actual irradiated power. As an additional check on relative power, a Nicolet FIR Fourier transform spectrometer was used to determine the spectral response of our InSb detector referenced to a pyroelectric detector. The Nicolet results cover a frequency range from 450 to 1650 GHz and are with-

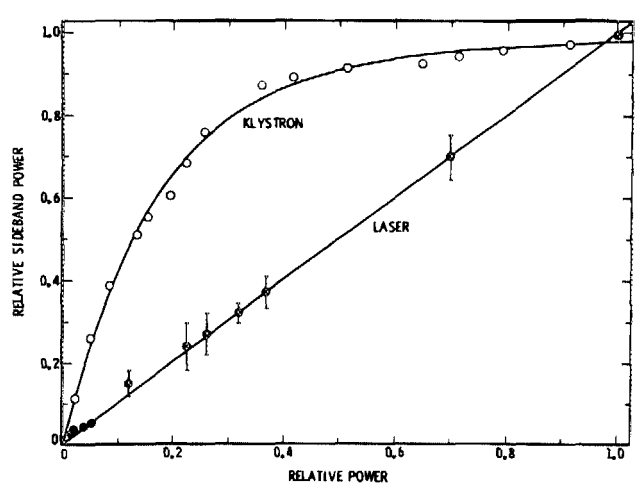

FIG. 2. Relative sideband power vs klystron power (O) and vs laser power (●) at 693-GHz center frequency.

in 3 to 10% of the four fixed point laser results. Therefore,the dominant error associated with all of our absolute power measurements is the $\pm$ 20% uncertainty of the Scientech power meter reading. Using the calibrated InSb detector the incident laser power at 693 GHz was measured to be 4 mW, and the laser sideband radiation at 600 and 786 GHz was measured to be 3.0 μW.

As an intermediate step to generating sidebands in the THz region, the FIR laser was operated at the 762-GHz formic acid line. With the entire setup unchanged, the upper and lower sidebands at 854 and 668 GHz were readily observed. The sideband characteristics were similar to the lower frequency results and were quite reproducible. In order to test the system at higher frequencies, the FIR laser was then operated on the difluoromethane line at 1627 GHz and the methanol line at 1839 GHz. Because of the rapid roll-off of the InSb detector at frequencies above 1000 GHz, a Ge bolometer cooled to 2.2 °K served as the detector for these measurements. The Schottky diode antenna was left the same as before, 1.7 mm long, due to the mechanical constraints of our existing corner cube structure. At these frequencies the antenna length was 9.2λ and 10.4λ, respectively. A reduction in coupling efficiency of the laser radiation was therefore expected. In addition, due to the shorter wavelength, the system was much less forgiving to instability and misalignment. The sidebands were, however, relatively easy to generate. Figure 3 shows a portion of a linear Fabry–Perot sweep verifying the generation of the 1839-GHz (163 μm) laser sidebands at 1746 (172 μm) and 1932 GHZ (155 μm). With the Ge bolometer calibrated against the InSb detector, we found that 22 mW of laser power at 1627 GHz produced about 2.5 μW of sideband power at 1532 GHz, while 3 mW of laser power generated approximately 100 nW of sideband power at 1932 GHz. The low efficiency is attributed mainly to the excessive length of the diode antenna.

To demonstrate the tunability and monochromaticity of the generated sideband radiation, several submillimeter rotational absorption lines have been measured with this system. The $2_{11} \rightarrow 2_{02}$ transition of HDO was detected in a 1.5-m path using the lower sideband of the 693–GHz formic acid laser line. Excellent signal-to-noise ratio were obtained by either video or lock-in detection. Our measured transition frequency of 599 926.81 MHz is in good agreement with that obtained by the harmonic generation of lower frequency klystrons.[16] We have also measured the $8_{17} \rightarrow 7_{16}$ and $11_{0,11} \rightarrow 10_{0,10}$ transitions of H_2CO near 600.3 and 786.3 GHz to verify the lower and upper sideband features seen in the Fabry–Perot interferometer sweeps. The major source of systematic error in these measurements is likely to be that in the assumed laser frequency. We have measured the laser frequency directly by using the corner cube as a harmonic mixer at 693 GHz and determined that the laser frequency varies by $\pm$ 0.5 MHz when the FIR cavity length is changed to produce a 10% drop in the output power level.

We have successfully developed a system which generates tunable laser sidebands in the submillimeter and far-infrared region and have demonstrated its potential for spectroscopic measurements. The system is more efficient than previously reported results even at high frequencies under adverse coupling conditions between the laser radiation and the Schottky diode mixer. The conversion efficiency ranges from about 0.075% at 693 GHz, where the coupling is optimum to about 0.003% at 1839 GHz, where the diode antenna is excessively long (10.4λ). This enhanced efficiency is in part due to the use of the Michelson interferometer to separate the sidebands from the carrier and in part due to the improved mount for the Schottky-barrier diode. With the numerous available FIR laser lines and the wide tunability ranges of klystrons, the high-resolution spectroscopic capabilities traditionally associated with microwave spectroscopy have been extended to the far-infrared region.

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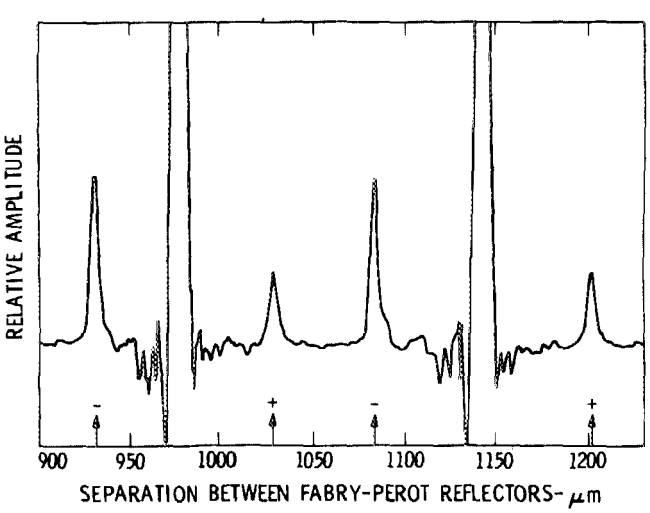

FIG. 3. Portion of the linear Fabry–Perot sweep at the 1839-GHz (163-μm) center frequency. The upper and lower sidebands at 1932 (155 μm) and 1746 GHz (172 μm) are marked by arrows and + and − signs, respectively. The horizontal scale is an absolute scale in μm showing the separation between the Fabry–Perot reflectors.

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