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LETTER TO THE EDITOR

Extension of the Range of Microwave Spectroscopy up to 1.3 THz

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At the present time, the upper frequency limit for high resolution scanning microwave studies using a phase-locked BWO oscillator as the radiation source (*1*) lies around 1 THz (*2*). The present upper frequency limit achieved by multiplication of the frequency of the primary radiation source also lies around 1 THz (*3*). This limit has not changed significantly during the last decade. However, the beat note between the third harmonic of a 600 GHz BWO and the 16th harmonic of a 78–118 GHz synthesizer using a Schottky diode has been observed during our development of a precise microwave broadband scanning spectrometer in the THz range (*2*). In addition, we have observed frequency harmonics produced by the BWO itself (*4*) during the first attempt at harmonic generation. In the present paper, we report the first results of frequency multiplication of submillimeter BWO radiation with a Schottky diode and the extension of the usable frequency range for microwave spectroscopy through 1.36 THz.

A block diagram of our experimental setup is presented in Fig. 1. The radiation of a submillimeter BWO operating in the 300 GHz frequency range and stabilized by a phase lock loop (*4*) was directed by a waveguide to the multiplier, as depicted in Fig. 2.A Schottky diode with capacitance 6–7 fF, resistance about 12 Ω, and upper frequency limit around 3 THz with regulated bias voltage was used as the nonlinear element. The radiation from the multiplier was directed through a spectroscopic acoustic cell commonly used in our experiments (*4, 5*) which was utilized here for harmonic selection and detection by observation of appropriate SO_2 spectral lines. The gas pressure in the cell was about 0.2 Torr. The lines were observed in the fundamental frequency of the BWO, which was not filtered out before entering the cell, as well as in the second, third, and fourth harmonics of the fundamental frequency.

The narrowness of the lines observed using the harmonics as opposed to the fundamental frequency, the proximity of the measured line frequencies to calculated ones in the well-studied SO_2 spectrum, and the relative intensities of the lines observed in groups all served as the main criteria for determining the correct harmonics. The dependence of line intensity on diode bias also served as an indicator of the proper harmonic. In particular, lines observed with the fundamental frequency had no measurable dependence on diode bias voltage. Examples of lines observed with harmonics under differing bias voltages are presented in Figs. 3–6. As may be seen from Fig. 4, optimal generation of third and fourth harmonic frequencies requires different bias voltages, i.e., different working points on the nonlinear diode characteristic.

The frequencies of lines measured using the BWO harmonics are listed in Table I along with calculated frequencies (*6*). The frequencies of several lines have been measured within an uncertainty of 300 kHz with the standard approach of tuning the phase-locked BWO frequency through the line center. The remainder of the lines were measured by scanning the phase-locked BWO with reference frequency marks. The differences between calculated and measured line frequencies are generally well within 1 MHz; lines measured on the shoulders of fundamental frequency lines show somewhat larger differences from calculated frequencies.

The results of the present work, in which molecular lines were observed with the second, third, and fourth harmonics of a BWO operating in the submillimeter-wave region, demonstrate an extension of microwave spectroscopy well beyond a frequency of 1 THz. In our opinion, a further significant extension of the frequency range can be achieved simply by using the methods described here with more sensitive receivers than the acoustic cell which was at our disposal. For example, receivers such as cryogenic bolometers similar to the InSb type (*2*) or even more sensitive bolometers with ^3He cooling or magnetic tuning can be used. With such receivers, the detection of higher harmonics than presented here can be achieved. Of course, use

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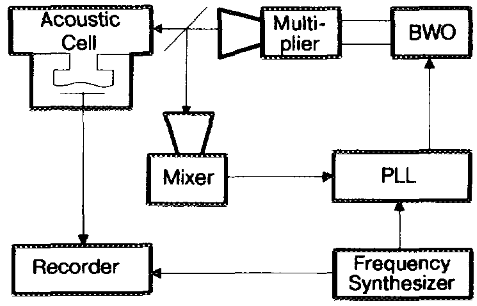

FIG. 1. A block diagram of the experimental apparatus.

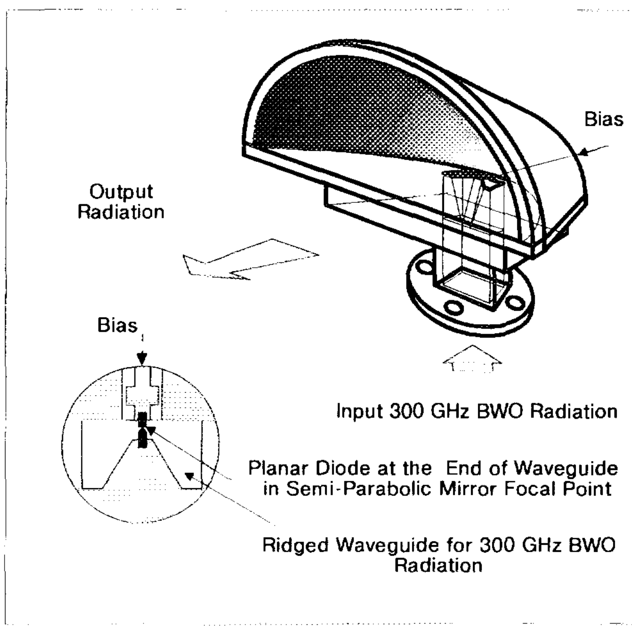

FIG. 2. Construction of the frequency multiplier on a planar Schottky diode.

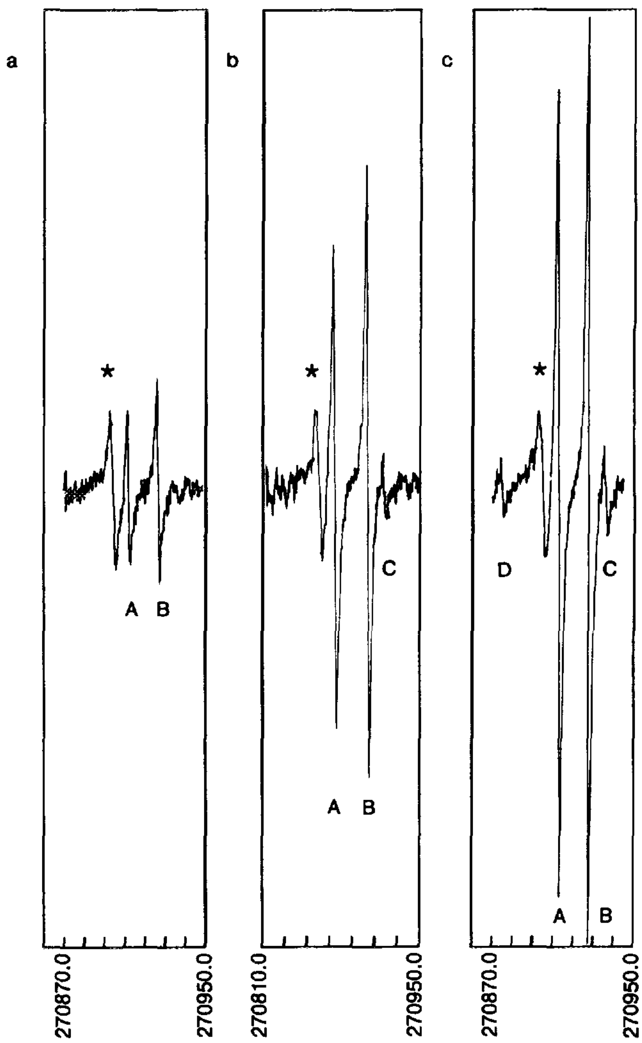

FIG. 3. Three traces of the same portion of the spectrum with different bias voltages demonstrating second harmonic generation. The bias voltages are (a) 0 V (diode shorted), (b) 0.3 V (diode current 0.5 mA), and (c) 0.5 V (diode current 2.8 mA). The fundamental BWO frequencies are shown in MHz. An unidentified SO_2 line observed with the fundamental BWO frequency is shown by an asterisk in all three traces. The second harmonic lines are distinguished by letters with the following frequencies and intensities:

A. $14_{3,11} \leftarrow 13_{2,12}$; $^{32}SO_2(000)$; 541 750.90 MHz; $1.1 \times 10^{-2}cm^{-1}$;

B. $30_{6,24} \leftarrow 30_{5,25}$; $^{32}SO_2(000)$; 541 810.56 MHz; 1.2×10^{-2} cm^{-1};

C. $13_{6,8} \leftarrow 13_{5,9}$; $^{34}SO_2(000)$; 541 844.24 MHz; $6.2 \times 10^{-4}cm^{-1}$;

D. $14_{6,8} \leftarrow 14_{5,9}$; $^{34}SO_2(000)$; 541 652.42 MHz; $6.6 \times 10^{-4}cm^{-1}$.

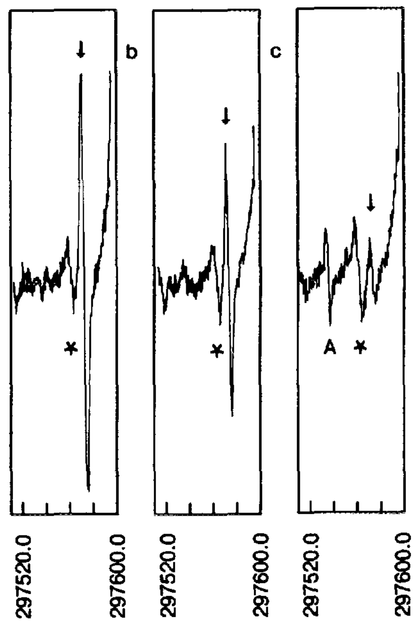

FIG. 4. Three traces of the same portion of the spectrum with different bias voltages demonstrating third and fourth harmonic generation. The bias voltages are (a) 0 V (diode shorted), (b) 0.2 V (diode current 3.3 mA), and (c) 0.4 V (diode current 5.0 mA). The fundamental BWO frequencies are shown in MHz. The $9_{3,7} \leftarrow 10_{0,10}$ (010) line of $^{32}SO_2$ at 297 571.35 MHz (intensity 1.7×10^{-6} cm^{-1}) observed using the BWO fundamental frequency is shown in all three traces with an asterisk. The $12_{7,5} \leftarrow 11_{6,6}$ line at 892 746.27 MHz (intensity 4.5×10^{-2} cm^{-1}), observed in the third harmonic, is designated by an arrow. The $17_{9,9} \leftarrow 16_{8,8}$ line at 1 190 182.09 MHz (intensity 6.6×10^{-2} cm^{-1}), observed in the fourth harmonic, is designated by the letter A in the third record. The lines are observed on the wing of the $^{32}SO_2$ (100) $24_{4,20} \leftarrow 24_{3,21}$ line at 297 612.83 MHz.

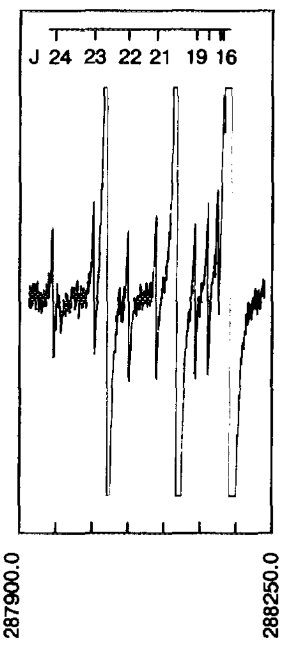

FIG. 5. A trace of the $K_a = 9 \leftarrow 8$ Q-branch of SO_2 near 864 GHz with J values from 16 to 24, observed in the third harmonic, demonstrates some scanning features of the multiplier. The fundamental BWO frequency is plotted in MHz. The very strong lines are seen in the fundamental BWO frequency.

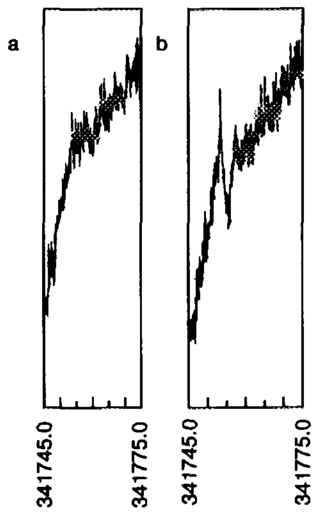

FIG. 6. Two traces of the 1.367 THz $21_{10,12} \leftarrow 20_{9,11}$ line of SO_2 (000) (intensity 6.9×10^{-2} cm^{-1}) are shown plotted against the fundamental BWO frequency. The line, observed in the fourth harmonic, is found on the wing of the fundamental $36_{5,31} \leftarrow 36_{4,32}$ (000) line at 341 673.98 MHz (intensity 4.3×10^{-2} cm^{-1}), which lies approximately 80 MHz away in fundamental frequency. The sign of the measured–calculated difference in Table I coincides with the sign of the wing frequency shift. Bias voltages: (a) 0.1 V (diode current 3.0 mA); (b) 0.4 V (diode current 5.6 mA).

TABLE I

Measured and Calculated Frequencies of $^{32}SO_2$ Ground Vibrational State Lines Observed in the Harmonics of the BWO Fundamental Frequency along with Number of Harmonics Used in Observation

SO_2 transition $J'_{K'_a,K'_c} \leftarrow J''_{K''_a,K''_c}$	Measured frequency MHz	Calculated frequency[1] MHz	M. − C. difference MHz	Harmonic number
$14_{3,11} \leftarrow 13_{2,12}$	541 751.2(20)	541 750.90	0.3	second
$30_{6,24} \leftarrow 30_{5,25}$	541 810.0(20)	541 810.56	-0.56	second
$24_{9,15} \leftarrow 24_{8,16}$	863 836.0(20)	863 835.61	0.39	third
$22_{9,13} \leftarrow 22_{8,14}$	864 145.51(30)	864 145.39	0.12	third
$11_{7,5} \leftarrow 10_{6,4}$	873 600.50(30)	873 600.57	-0.07	third
$12_{7,5} \leftarrow 11_{6,6}$	892 745.5(20)	892 746.27	-0.77	third
$10_{8,2} \leftarrow 9_{7,3}$	955 436.8(20)	955 437.04	-0.24	third
$12_{8,4} \leftarrow 11_{7,5}$	993 766.22(10)	993 766.25	-0.03	third
$10_{10,0} \leftarrow 9_{9,1}$	1 156 015.8(20)	1 156 016.10	-0.3	fourth
$17_{9,9} \leftarrow 16_{8,8}$	1 190 181.4(20)	1 190 182.09	-0.69	fourth
$11_{11,1} \leftarrow 10_{10,0}$	1 274 700.34(30)	1 274 700.58	-0.24	fourth
$13_{11,3} \leftarrow 12_{10,2}$	1 313 099.0(20)	1 313 098.86	0.14	fourth
$21_{10,12} \leftarrow 20_{9,11}$	1 367 017.4(20)	1 367 018.75	-1.35	fourth

[1] Values calculated from Ref. (6).

of a bolometer instead of an acoustic cell demands harmonic selection before the bolometer by some monochromator, such as the well-known FIR échelle grating.

Another approach to higher frequency generation is to utilize frequency BWOs as the primary sources for harmonic generation. In addition, an increase of the power radiated in the harmonics can be expected by suitable construction of a planar Schottky diode with additional antennae connected to each end of the diode to form a half-wavelength dipole for the desired harmonic.

In conclusion, we have demonstrated the successful use of a Schottky diode for harmonic generation of submillimeter BWO radiation and set a new upper limit for the frequencies used in microwave spectroscopy. We have also discussed several methods of extending the range of microwave spectroscopy to still higher frequencies.

ACKNOWLEDGMENTS

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