

## Spectroscopy of the Harmonics of Submillimeter Backward Wave Oscillators

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The harmonics content in the radiation generated by a submillimeter backward wave oscillator is described for the first time. Lines were observed in the second and third harmonic using an acoustic cell as a receiver in the 600-GHz frequency range. A possible explanation is given for the harmonic generation. It is thought that strong enough electron bunching induces nonsinusoidal convection currents in slow-wave systems. This is analogous to frequency multiplication in centimeter-wave klystrons. Possible means for reaching higher frequencies using more sensitive receivers such as cryogenic bolometers and changing construction and/or operational regimes of oscillators are pointed out. © 1995 Academic Press, Inc.

### 1. INTRODUCTION

This paper is a continuation of a recent experiment for sweeping a frequency-locked backward wave oscillator (BWO) up to the 1-THz frequency range (1). During that experiment, beats between the third harmonic of a BWO of the type in Ref. (2) and the sixteenth harmonic of a millimeter-wave synthesizer were observed at 1.7 THz on the planar Schottky diode in the multiplier-mixer used. That observation served as the starting point for the present work, in which we attempted to produce BWO frequency multiplication in the common way—by using a nonlinear element, in this case the same planar Schottky diode as used in Ref. (1). Because a sensitive helium-cooled InSb bolometer was not at our disposal at Nizhnii Novgorod, we used observation of the spectral lines at the proper frequencies in the acoustic cell used commonly in our experiments (see, e.g., Ref. (3) for harmonic detection. We started with an OB-30 tube working in a longer submillimeter region as with the more powerful one.

### 2. EXPERIMENTAL DETAILS

We started from a common scheme for such experiments: the radiation from the BWO was directed by a waveguide to the multiplier, and the output of the multiplier was directed by a horn to the acoustic cell. The output signal from the multiplier was not filtered from the fundamental BWO frequency; the smaller part of the output containing the fundamental frequency was directed by a beam splitter to the mixer of the BWO phase lock loop (PLL). We do not describe the multiplier here for reasons that will be clear from the following. A block diagram of the experimental setup is presented in Fig. 1a.

In these experiments we observed spectral lines using the second harmonic of a BWO. For example, in Fig. 2a the  $J = 1-0$  line of  $^{14}\text{NH}_3$  was observed at 572 499 MHz when the BWO was phase-locked to 286 249 MHz. For testing the reproducibility of the effect, the ammonia sample was replaced with a sample enriched with  $^{15}\text{NH}_3$ . A spectral line at 572 112 MHz corresponding to the  $J = 1-0$  transition of  $^{15}\text{NH}_3$

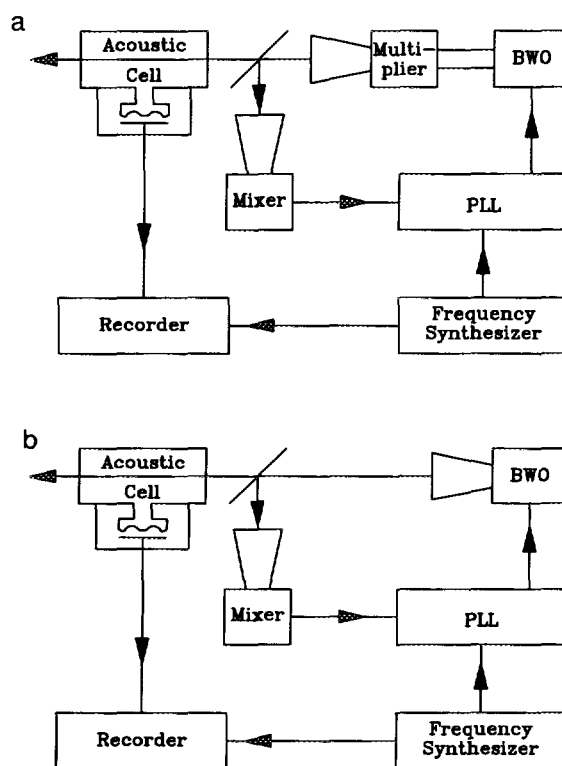


FIG. 1. Block diagram of the experimental setup: (a) initial block diagram including the multiplier; (b) final block diagram of setup used in observation of lines in BWO harmonics (multiplier removed).

appeared (see Fig. 2b) when the BWO was tuned by a lock-in loop to half this frequency. Additionally, the widths of both lines were half of what would be expected if they were observed that the BWO fundamental frequency.

We first supposed that the second harmonic radiation came from the multiplier, but after some unsuccessful attempts to increase the harmonic power by tuning the multiplier, we simply removed it and directed the BWO radiation straight into the cell and continued to observe lines in the second harmonic of the BWO frequency (see the block diagram in Fig. 1b).

Additional observations of spectral lines in harmonics of a lower frequency OB-24 BWO tube have been made without a multiplier between the BWO and the cell.

In Fig. 3 the whole  $K$  structure of the  $J = 11-10$  transition of  $\text{CH}_3\text{F}$  can be seen; this transition is situated near 561 GHz but is observed with a BWO whose fundamental frequency range is 187 GHz.

Figure 4 shows the  $K_a = 5-6$  transitions of the  $^{32}\text{SO}_2$   $Q$  branch. These transitions lie between the much stronger (in the spectrum)  $^{34}\text{SO}_2$  ground state lines at the fundamental frequency of the BWO (again nearly 187 GHz), but the frequency of the  $K_a = 5-6$   $Q$  branch for  $^{32}\text{SO}_2$  is about 561 GHz. Measurements of  $Q$  - branch line frequencies by a frequency synthesizer were (after multiplication by 3) in agreement with theory for  $K_a = 5-6$  transitions of the  $Q$  branch to within 50 kHz. This  $Q$  branch was also observed in the third harmonic of the BWO used. This result makes it possible to estimate the intensity of the third harmonic compared to the fundamental frequency.

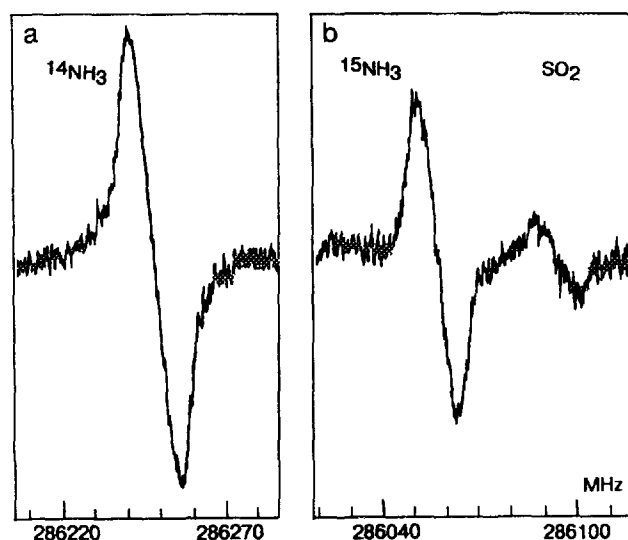


FIG. 2. Spectra of the  $J = 1-0$  lines of  $^{14}\text{NH}_3$  (a) at a frequency of 572 499 MHz and (b) at a frequency of 572 112 MHz obtained in the second harmonic of the BWO used (see the frequency scale in the figures). The sensitivity of the spectrometer in the fundamental frequency in the vicinity of the lines was on the order of  $10^{-8} \text{ cm}^{-1}$ . Note that ammonia lines are approximately two times narrower than the line due to the impurity of  $\text{SO}_2$  in (b) observed in the fundamental frequency due to the increase of the frequency deviation in the second harmonic.

Namely, lines with intensities of  $10^{-2} \text{ cm}^{-1}$  in the third harmonic look much smaller than lines with intensities from  $10^{-4}$  to  $10^{-5} \text{ cm}^{-1}$  in the fundamental frequency; numerical estimation gives a power reduction by a factor of  $3 \times 10^4$ .

A number of  $^{32}\text{SO}_2$  lines in the ground and excited states were observed using the second harmonic of an OB-24 BWO tube (from the  $^{32}\text{SO}_2$  (000)  $21_{-16}-21_{-14}$  line at 363 159.251 MHz to the  $^{32}\text{SO}_2$  (000)  $11_{-10}-12_{-8}$  line at 463 011.442 MHz). We

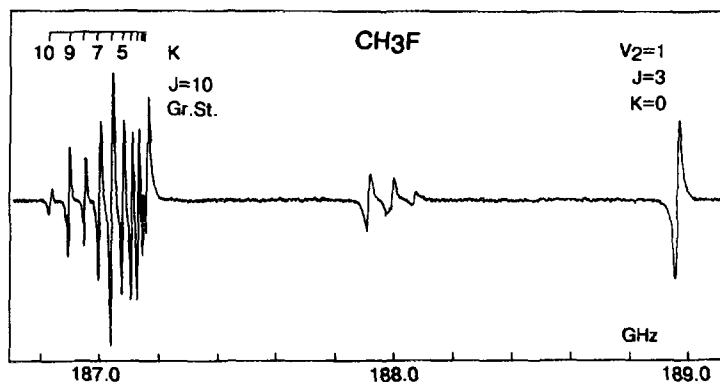


FIG. 3. The  $K$  group of the  $J = 11-10$  rotational transition of the ground state of  $\text{CH}_3\text{F}$  at 561 GHz observed in the third harmonic of the BWO used (see the frequency scale in the figure).  $K$  values are shown in the figure; note the increased intensity of lines with  $K = \text{multiples of three}$ . Note the narrowness of these lines compared with the excited state  $\text{CH}_3\text{F}$  line observed in the same figure in the fundamental frequency (to the right;  $\nu = 188\,962 \text{ MHz}$ ,  $J = 4-3$ ,  $K = 0$ ,  $v_2 = 1$ ) and with the unknown lines in the center.

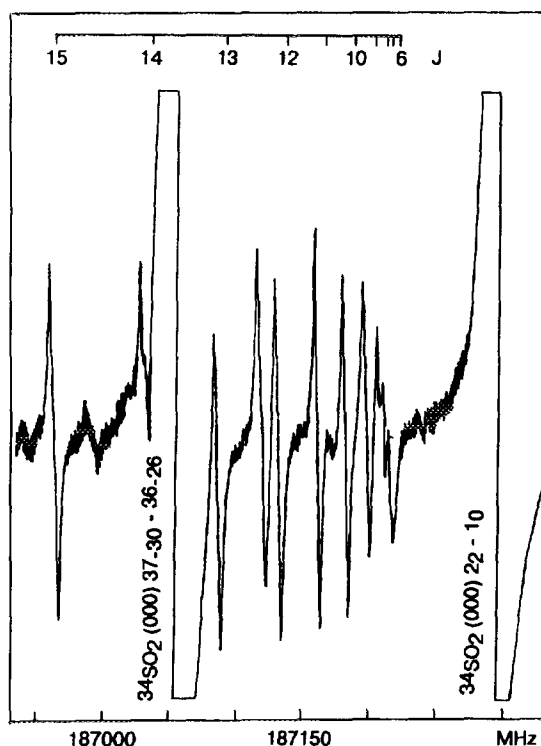


FIG. 4.  $Q$  branch of the  $^{32}\text{SO}_2$   $K_a = 5-6$  transition ( $J$  values marked in the figure) around 561 GHz observed in the third harmonic of the BWO used (see the frequency scale in the figure) between the ground state lines of  $^{34}\text{SO}_2$  in natural abundance observed in the fundamental BWO frequency (lines limited by the recorder). The intensity of the  $Q$ -branch lines around  $10^{-2} \text{ cm}^{-1}$  and the intensities of lines in the fundamental frequency were for  $37_{-30}-36_{-26}$ ,  $1.9 \times 10^{-4} \text{ cm}^{-1}$ , and for  $22_{-10}$ ,  $3.1 \times 10^{-5} \text{ cm}^{-1}$ . Measured in this way, frequencies of  $Q$ -branch lines coincide with those calculated in Ref. (5) to within 50 kHz (see Table I).

estimated a reduction of power on the second harmonic compared to the power of the fundamental frequency by a factor of  $3 \times 10^3$ .

Table I contains frequencies measured in the BWO second and third harmonics of  $\text{SO}_2$  lines together with those calculated from Ref. (5). We did not include frequencies of lines which were observed on the wings of stronger lines as can be seen in Fig. 4.

These effects have previously been attributed to the side effects of frequency multiplication on the nonlinear element always present in every system using BWO frequency stabilization (in our case a planar Schottky diode in the side arm). But in every case only a small part of the BWO power was used for frequency stabilization, i.e., could participate in the frequency multiplication process. But in the present work the full power of a BWO was directed into the multiplier, producing currents through its Schottky diode on the order of 2–3 mA. When removal of the multiplier from the main setup in the present experiment did not produce a change in the signal from the spectral line observed in the BWO harmonic it became clear that an explanation of the previous observations based on side effects of multiplication produced by a small amount of BWO power could not be correct.

TABLE I  
Measured and Calculated Frequencies of Some SO<sub>2</sub> Lines  
Observed in Harmonics of Backward Wave Oscillators

<sup>32</sup> SO <sub>2</sub> transition $J'_{K'_1, K'_2} \leftarrow J''_{K''_1, K''_2}$	Measured frequency MHz	Calculated frequency <sup>1</sup> MHz	M. - C. difference kHz	BWO harmonic number
21 <sub>4,18</sub> ← 21 <sub>3,19</sub>	363 159.251	363 159.260	-9	second
18 <sub>5,13</sub> ← 18 <sub>4,14</sub>	455 348.276	455 348.222	54	second
19 <sub>5,15</sub> ← 19 <sub>4,16</sub>	456 352.344	456 352.309	35	second
16 <sub>5,11</sub> ← 16 <sub>4,12</sub>	457 315.484	457 315.489	-5	second
12 <sub>2,10</sub> ← 11 <sub>1,11</sub>	463 011.442	463 011.409	33	second
16 <sub>6,10</sub> ← 16 <sub>5,11</sub>	560 613.573	560 613.521	52	third
15 <sub>6,10</sub> ← 15 <sub>5,11</sub>	560 891.025	560 890.972	-53	third
11 <sub>6,6</sub> ← 11 <sub>5,7</sub>	561 490.484	561 490.522	-38	third

### 3. DISCUSSION

Analogously, in our opinion, the effect observed at lower frequencies is multiplication of the frequency in two-cavity klystrons (see, e.g. Ref. (4)) when bunching of electrons in the beam produced by the input signal in the first cavity is strong enough. Then electron pulses in the second cavity induce current containing not only the fundamental (input) frequency component but also its harmonics. Tuning the second cavity to the desired harmonic frequency then permits strong enough output signal in the harmonic to be obtained; in this manner frequency multiplication by more than 10 times has been achieved (4). But, to the best of our knowledge, such an effect has never been mentioned in connection with submillimeter backward wave oscillators.

We suggest that the electron beam bunching in the BWO was strong enough to induce harmonics of the fundamental frequency of BWO oscillation in the slow-wave structure, and some amount of the power in these harmonics found its way out of the tube. If so, some interesting consequences may result even if the level of these harmonics is very low compared to our previous estimates. First, these harmonics can still serve as a useful source of coherent radiation when modern high-sensitivity bolometers, which are much more sensitive than room-temperature acoustic detectors, are used as receivers.<sup>1</sup> Second, these harmonics may be an even more interesting source of higher frequencies, keeping in mind that at least for lower frequencies the intensity of the harmonics produced in such a manner decreases very slowly with the number of harmonics (4). So if lines in the second and third harmonics were observed with an acoustical cell, it could be very interesting to follow these and higher harmonics using the more sensitive bolometers as receivers. We could not view harmonics at frequencies higher than approximately 600 GHz using an acoustical cell because of the lack of sensitivity.

Our experiments also used acoustic detection for harmonic selection (e.g., ignoring strong fundamental spectral components if there were no absorption lines at the fun-

<sup>1</sup> For example, that used in Ref. (1) or newly developed by the QMC Co. using magnetic resonance or <sup>3</sup>He cooling.

damental frequency). For harmonic selection in the bolometer case, common far-infrared échelle or other monochromator techniques can be used.

So by continuing work on "common" BWO frequency multiplication on high-frequency diodes, one encounters the possibility of extending the range of coherent microwave spectroscopy in the manner described in this short communication; moreover, it is quite possible that, keeping in mind this newly presented information, one may attempt to change the construction and/or operational regimes of the submillimeter BWOs to enhance the process of harmonic generation in them.

Due to the nature of the effect, it can be expected to occur more easily in the more powerful tubes, producing stronger bunching of the electrons, so demand for more powerful tubes for more effective multiplication is common for both types of frequency multiplication considered. We plan to continue such studies jointly with I. Physical Institute of the University of Cologne and ISTOK Research and Development Co. (which developed and produced the tubes mentioned here).

With this short communication we have presented a new possibility for overcoming the present frequency limitation of BWO-based microwave methods and, it is hoped, increasingly upper frequency of such precise broadband scanning coherent spectroscopy well beyond the existing terahertz limit.

#### ACKNOWLEDGMENTS

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