Extension of the range of microwave spectroscopy to 1.5 THz

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The frequency range of microwave spectroscopy has been extended to 1.5 THz through multiplication of the frequency of a 300-GHz submillimeter backward-wave tube to the fifth harmonic with the help of a new frequency multiplier with a planar Schottky diode. A spectroscopic cell with an acoustic detector was used to select and detect harmonics through observation of spectral lines of gases. Lines of SO_2 over the range 541–1524 GHz were observed at the second, third, fourth, and fifth harmonics of the fundamental frequency of the backward-wave tube. Ways to expand the range of microwave spectroscopy substantially further are pointed out. © *1995 American Institute of Physics.*

Expanding the frequency range is one of the basic directions in the development of microwave spectroscopy. The standard approach has been to multiply the frequency of sources of centimeter- and millimeter-range radiation through the use of a nonlinear element, usually a semiconductor point-contact diode. That approach was first taken by W. Gordy, who advanced into the long-wave part of the submillimeter range back in 1954 (Ref. 1). The use of primary sources of submillimeter radiation—backward-wave tubes of the type in Ref. 2—for microwave spectroscopy allowed us to exceed, in 1973, the upper frequency limit on microwave research using harmonic generators. A frequency of 874 GHz was reached,[3] in comparison with the 813 GHz in Ref. 4. The upper frequency limits were later pushed upward to 1100 GHz in spectroscopy with submillimeter backward-wave tubes (Ref. 5, for example) and to 1036 GHz in spectroscopy with frequency multiplication of millimeter-range sources (Ref. 6, for example). These remained the limits for more than a decade.

In this letter we are reporting an extension of the frequency range of microwave spectroscopy to 1524 GHz. This advance has been achieved through frequency multiplication of (we wish to emphasize this point) a 300-GHz submillimeter backward-wave tube. The radiation from the submillimeter backward-wave tube, whose frequency was stabilized by a phase self-adjustment system, was directed along a waveguide to a frequency multiplier which we have developed. This multiplier is based on a mixer involving harmonics.[7] A planar Schottky diode with a capacitance of 0.007 pf and a spreading resistance of 10 Ω is used in the multiplier. An adjustable dc voltage is applied to the diode.

The radiation from the output of the multiplier, which contains power at both the fundamental frequency and harmonics of the backward-wave tube, is directed by a semiparabolic mirror to the spectroscopic cell, which is equipped with the acoustic detector which we have customarily used for spectroscopy (see Ref. 5 for the procedure).

For the observations we selected the well-known rotational spectrum of the SO_2 molecule, which has long been used as a reference.[3] Extensive tables of the spectral lines of SO_2 have been calculated up to frequencies on the order of several terahertz on the basis of the data of Ref. 8.

The lines observed at the harmonics are narrower in the spectrum, because the frequency changes more rapidly at the harmonics than at the fundamental frequency of the backward-wave tube. The intensities of the lines observed at the harmonics depend strongly on the bias voltage on the diode, which determines the position of the working point on the current–voltage characteristic of the diode. The intensities of the lines observed at the fundamental frequency, in contrast, are not visibly dependent on the bias voltage. These characteristics, along with the agreement of the measured and tabulated line frequencies within a relative error of 10^{-6}–10^{-7}, were utilized to identify the spectral lines which were observed. A total of more than 30 spectral lines were observed and identified at the second, third, fourth, and fifth harmonics of the frequency of the

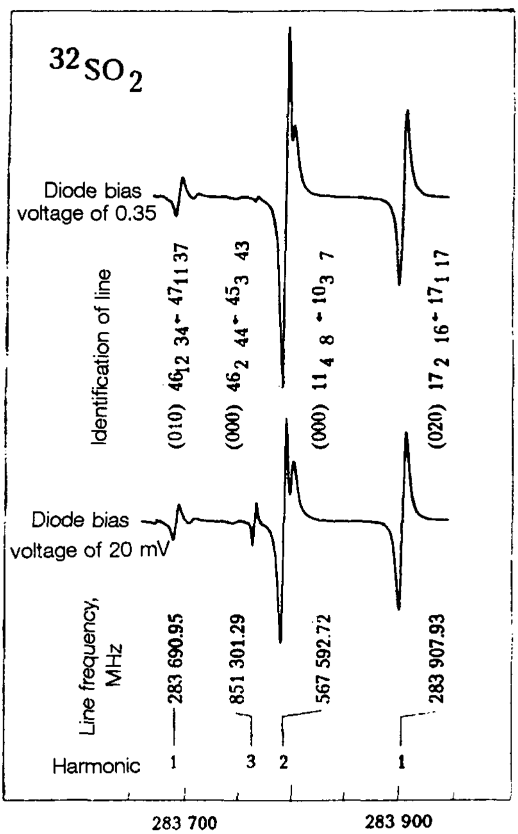

FIG. 1. Part of the spectrum of SO_2. Differences can be seen in the behavior of the intensity as a function of the bias voltage on the Schottky diode and in the widths of the lines observed at the first, second, and third harmonics of the backward-wave tube.

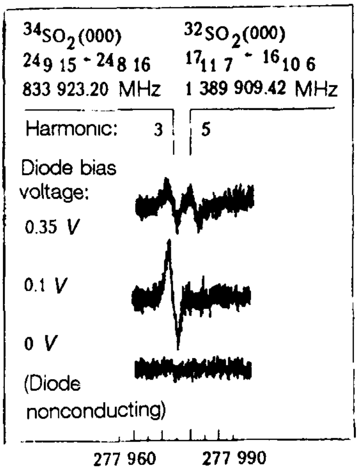

FIG. 2. Part of the SO_2 spectrum with lines observed at the third and fifth harmonics of the backward-wave tube.

backward-wave tube. Only a few of these lines are shown here, to save space in this letter.

Figures 1–3 show parts of the SO_2 spectrum, the frequency of backward-wave tube in megahertz (as shown on the scale), the bias voltage on the Schottky diode, the identification of the observed lines in the ordinary asymmetric-top notation,[6] the corresponding tabulated frequencies, and the numbers of the harmonic at which the line was observed (1 means the fundamental frequency of the backward-wave tube).

The spectra in Figs. 1 and 2 demonstrate the behavior mentioned above. Figure 3 shows the line observed at 1 524 236.16 MHz, which is the highest frequency observed to date for microwave spectroscopy.

We believe that a further, and extremely substantial broadening of the frequency range of microwave spectroscopy can be achieved simply by using a more sensitive detector along with the combination (described in this letter) of a submillimeter backward-wave tube with a Schottky-diode frequency multiplier, instead of the thermal acoustic detector which operates at room temperature and which was the only one at our

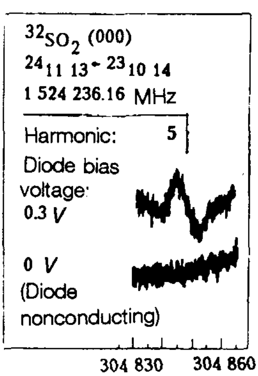

FIG. 3. Line of SO_2 observed at the fifth harmonic of the backward-wave tube, at the highest frequency to date for microwave spectroscopy, 1 524 236.16 MHz.

disposal. As this detector one might use, for example, the InSb bolometer operating at liquid-helium temperature which was used in Ref. 7 or even more-sensitive bolometers cooled with ^{3}He and/or with magnetic tuning to the region above 1 THz.

We are planning experiments on frequency multiplication in which higher-frequency backward-wave tubes will be used as the primary sources. We also plan to study the possibility of improving the emission of harmonics by the planar Schottky diode which generates them. One possibility here is to equip the diode with a pair of antennas to form a half-wave vibrator for emission of the harmonic of interest.

In summary, the results of this study on frequency multiplication of a submillimeter backward-wave tube, in which spectral lines have been observed at the second, third, fourth, and fifth harmonics of the tube, demonstrate an expansion of the frequency range of microwave spectroscopy by a factor of about 1.5 in comparison with the previous record. These results point out ways to expand the frequency range of microwave spectroscopy substantially further.

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