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LETTER TO THE EDITOR

Precision Broadband Spectroscopy in the Terahertz Region

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For the first time frequency and phase stabilization of continuously tunable Backward Wave Oscillators (BWOs) at frequencies up to 1 THz have been achieved, introducing high-resolution scanning spectroscopy with microwave accuracy into the terahertz region. A precision tunable 78- to 118-GHz frequency synthesizer and a newly constructed broadband multiplier-mixer ensured reference signals over hundreds of GHz. This technique, combined with a sensitive He-cooled InSb detector, extends precision broadband spectroscopy into the entire region covered by high-frequency BWOs, probably up to 1.5 THz. The rotational spectra of the simple four-atomic molecule HSSH and its various isotopomers are particularly amenable to broadband spectroscopy and are used to demonstrate the wide tunability of the BWOs.

High-resolution rotational spectroscopy in the submillimeter-wave region has been achieved up to now mainly by frequency multiplication of lower frequency sources (see, e.g., Refs. (*1, 2*)) or by the use of BWOs as primary sources (*3*). Both methods, however, suffer from one serious limitation, namely that neither of them could be used for precision, continuous broadband spectroscopy ($\geqslant$5 GHz) in the terahertz region.

This situation has changed very recently with the introduction of high-frequency BWOs ($\geqslant$400 GHz) in combination with precisely and continuously tunable frequency synthesizers (e.g., 78–118 GHz) which deliver sufficient power for frequency multiplication (*4*). At the laboratory of the Institute for Applied Physics, Nizhnii Novgorod, we have developed a broadband multiplier-mixer based on the planar Schottky technique, together with the phase-lock electronics similar to the one described in Ref. (*4*), to which a HEMT IF preamplifier was added. Introduction of these techniques into the existing Cologne spectrometer in Germany (*5*) led to the high-precision broadband phase-locked spectroscopy up to about 1 THz which is the subject of the present Letter.

In these experiments we have employed BWO tubes up to 1 THz supplied by the ISTOK Research and Production Co. located in Fryazino, near Moscow. We have now succeeded for the first time to frequency and phase stabilize high-frequency BWOs to the harmonic output of either our Gunn oscillators or a 78- to 118-GHz frequency synthesizer, supplied by the Institute of Electronic Measurement, KVARZ, Nizhnii Novgorod. Both sources provide stabilized sweep outputs. Although the experimental arrangement will be discussed in more detail elsewhere, it will suffice here to show in Fig. 1 a schematic diagram of our present spectrometer setup. The crucial parts are the high-frequency and broadband tunable BWO, the newly constructed multiplier-mixer, the tunable millimeter-wave frequency synthesizer, and the He-cooled InSb detector. The power delivered by the BWOs depends on the operating frequency but ranges typically between 10 and 1 mW for the frequency range 300 GHz to 1 THz, respectively. The multiplier-mixer consists of a planar Schottky diode, mounted at the end of a grooved millimeter waveguide. The diode is placed in the focal point of a semiparabolic mirror illuminated with the submillimeter radiation. Once the device is properly adjusted, no further tuning is required. The stepwidth of the synthesizer can be as low as 100 Hz, with a leveled power output of 6 mW and peak values of 12 mW. The synthesizer and the data acquisition are PC controlled. Continuous scanning over the entire 40 GHz of the synthesizer results in hundreds of GHz at the BWO frequency. This way we have achieved continuous beat notes sufficient for a 200-GHz scanning of the BWO, locked at the fifth harmonic of the synthesizer.

The broad range tunability of the BWOs has been demonstrated by us by producing wide spectral scans in the terahertz region (*5, 6*). Equally impressive on these BWOs is their relatively constant power output

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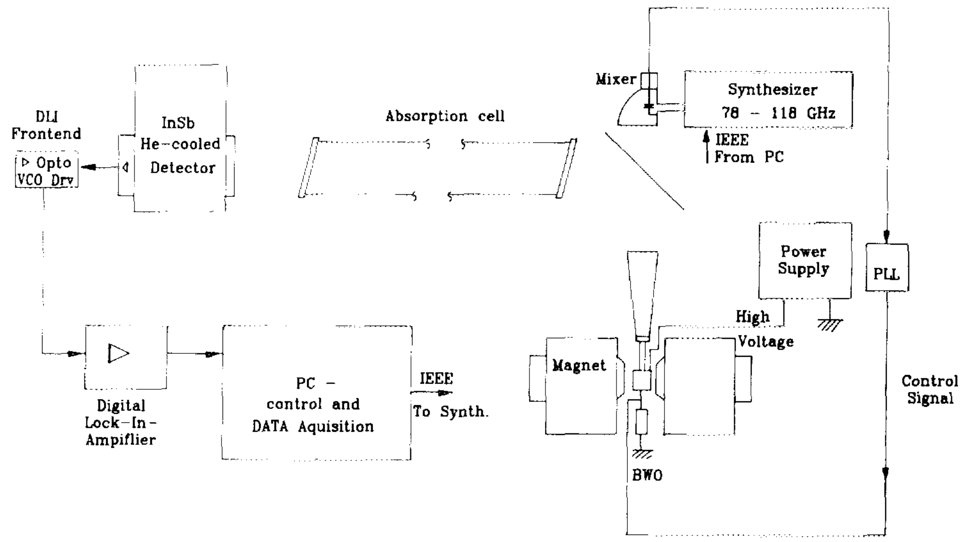


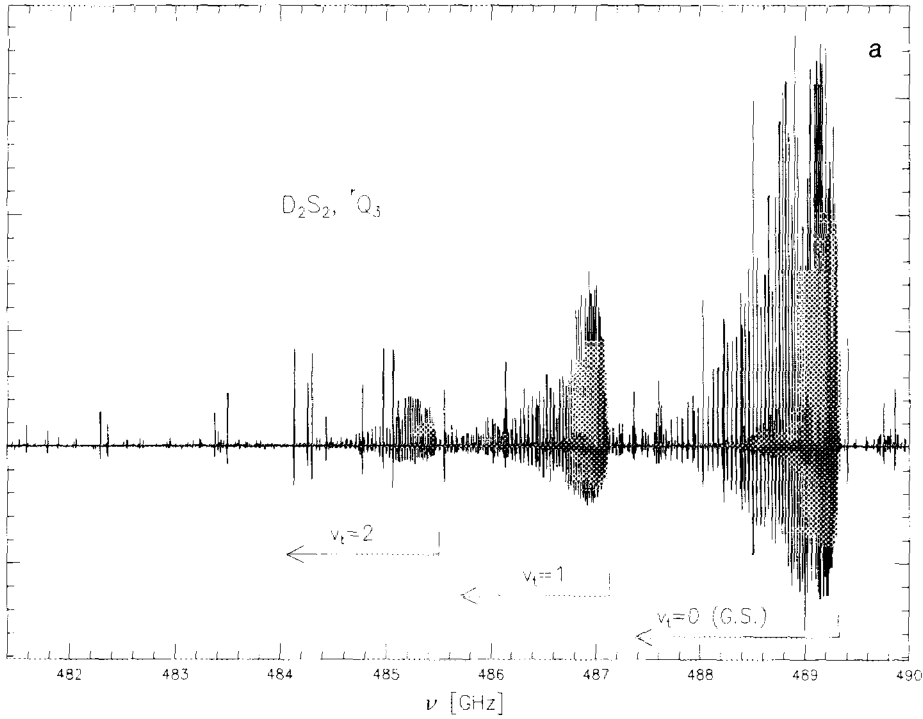
FIG. 1. Schematic diagram of the Cologne terahertz spectrometer in Germany. The backward wave oscillator (BWO) is controlled by a phase-lock loop. The radiation from the BWO passes a beam splitter. A portion of the power is mixed with a harmonic of a continuously tunable synthesizer to provide the lock signal. In some experiments, the synthesizer is replaced by a Gunn oscillator.

over broad frequency regions. These two properties, wide tunability and fairly constant power output, make them presently superior to any other monochromatic source we have used so far. In the following we present some recent spectral recordings obtained with several different BWOs operating between 300 GHz and 1 THz in order to demonstrate precision broadband spectroscopy.

As an example of the relatively constant power output, we presented an 8-GHz wide scan of the $^rQ_3$ branch of HSSH near 979 GHz, which has been recorded in the free running mode of the BWO (*5*). The Boltzmann population distribution over the rotational energy levels is displayed beautifully after subtraction of a baseline, providing direct evidence for the constant power output of the tube.

As a typical example of an extremely wide frequency- and phase-stabilized scan, we show in Fig. 2 a recording of the $^rQ_3$ branch of DSSD at 489.2 GHz, covering approximately 8.5 GHz. To our knowledge a scan of that frequency range has never been achieved before, where its width has been limited only by the adjustment of the high voltage of the BWO's power supply. Here we see for the first time two torsionally excited states of DSSD clearly displaced toward lower frequencies from the ground state. With the asymmetry parameter $\kappa = -0.999\,999\,315\,1(57)$, very close to -1, the symmetric-top limit, the individual J lines are very tightly packed, leading to the extremely compact Q branches. The result is that for DSSD the individual Q-branch patterns of the torsionally excited states do not overlap with each other and are clearly discernible. In Fig. 2a the location of the Q branches in the ground state and the first and second excited torsional states are marked, with the bandhead located at the high-frequency side. Although we will discuss the details of the spectrum and its assignment elsewhere, we present in Fig. 2b a somewhat higher spectral resolution recording of the ground state Q branch, and in Fig. 2c we zoom in on the bandhead. This example serves as a fine demonstration of the sensitivity of the spectrometer, the quality of the spectra, and the information content one can expect from broadband scanning in the submillimeter region. In Fig. 3 we include a recording of the $^rQ_4$ branch of DSSD at 629 GHz in the vibrational ground state. One notes that the intensity envelope of the individual J transitions follows approximately a Boltzmann distribution. In addition, one recognizes two additional Q branches interwoven with the ground state but exhibiting only about 10% of its intensity: they belong to $DS^{34}SD$ and the first excited stretching state. A high-resolution recording of these Q branches shows that the K-doubling is not resolvable for $J < 55$. But for larger J values the K-doubling is resolved, shows the typical nuclear spin statistical weights of 6:3, and is anomalous, i.e., the order of the $K_a = 3$ levels is inverted.

For the lower Q branches (i.e., $K_a = 2, 3$) the K-doubling is already resolved for low-J values, resulting in a fairly complex structure which has been unraveled by Winnewisser and Helminger (*7*). For reasons of



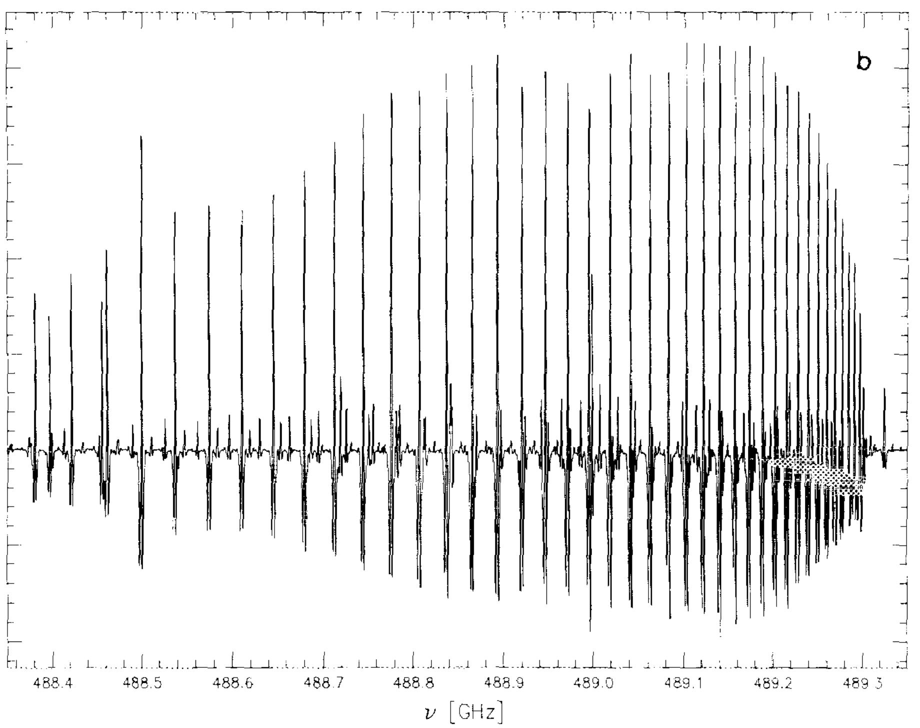


FIG. 2. A single broad scan of 8-GHz width of the $^rQ_3$ branch of DSSD with different frequency resolution. Each scan consists of 6000 points. (a) The stepwidth per point amounts to 1.25 MHz. The intensity variations result from frequency dilution. (b, c) The stepwidth is 0.30 MHz. The Q branch is degraded to lower frequencies. The bandhead is close to 489.3 MHz. (c) is a zoom of the bandhead showing the J assignment. The weaker lines are from $DS^{34}SD$.

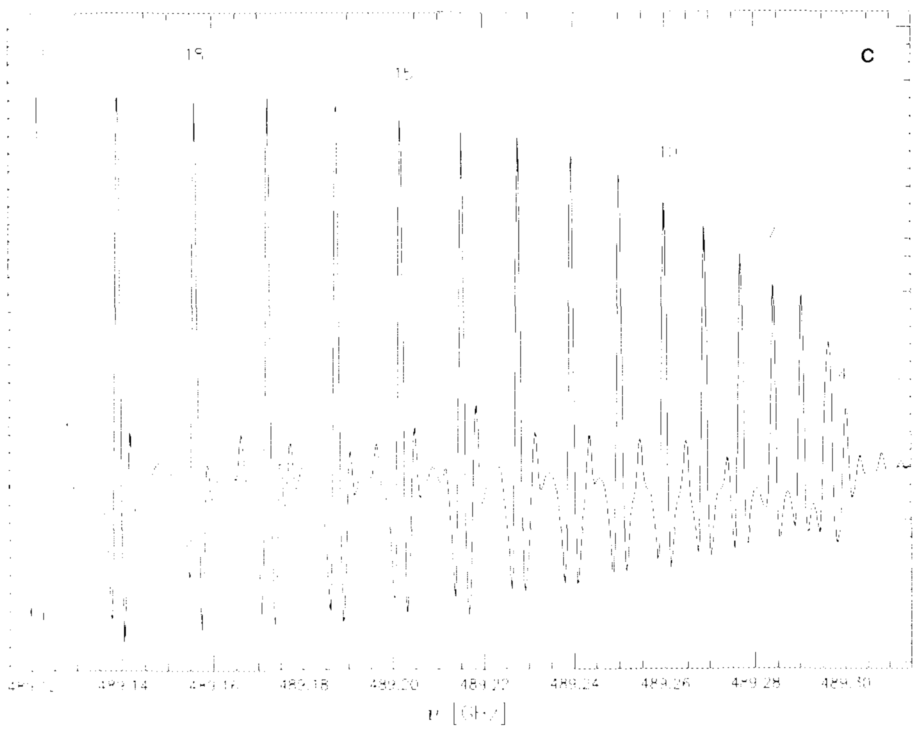


FIG. 2—*Continued*

comparison with the very early recorder tracing of the $^{r}Q_2$ branch of DSSD, we show in Fig. 4 a very recent scan over the same frequency region. In comparing the two spectra, i.e., Fig. 4 here and Fig. 6 of Ref. (*7*), one notes that aside from the somewhat improved resolution, the new recording contains for the ground state essentially the same information, but considerable new information is revealed for various excited vibrational states and for different isotopomers.

Finally, we present in Fig. 5 a recording of the 9_{81}–9_{72} transition of H_2S at 973.85 GHz. This measurement is part of an ongoing program to provide spectroscopic data of astrophysically relevant molecules in the terahertz region.

It should be pointed out that the continuous tunability of the BWOs covers almost one octave. In summary, we have achieved for the first time with BWOs frequency- and phase-locked performance with hitherto unknown wide tuning capabilities into the terahertz region. The BWOs tested in the present experiments are of superb quality and furnish a real alternative to the commonly used frequency-multiplication techniques. In fact, aside from the tuning capability, the output power of the tubes increases the hope that frequency multiplication of their output may allow spectroscopy up to 2 THz. The first piece of evidence for the feasibility of such an idea comes from our detection of beat notes on the mixer at 1.7 THz by using the 16th harmonic of the frequency synthesizer at 106.6 GHz and the 3rd harmonic of the BWO at 568 GHz, presented in Fig. 6.

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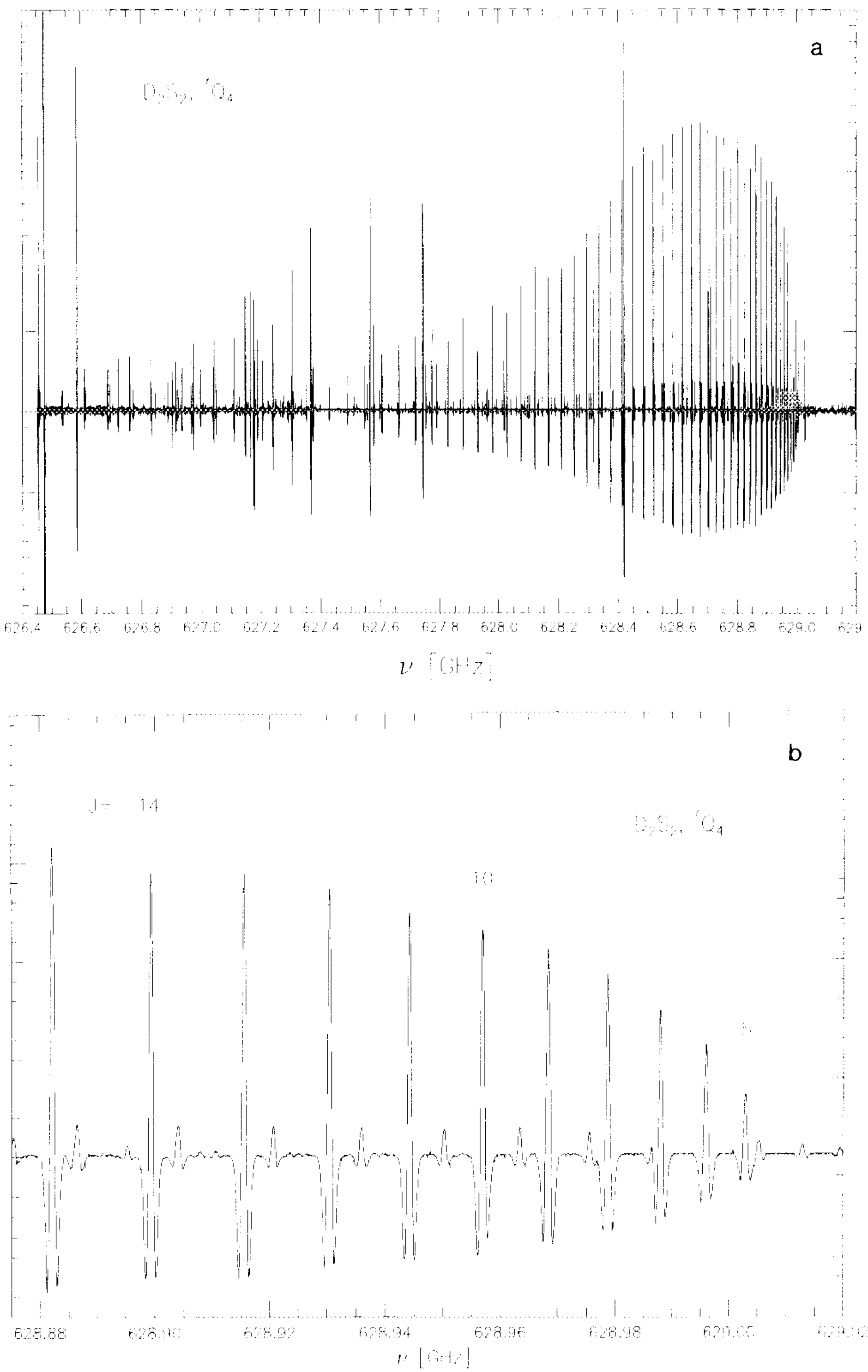

FIG. 3. (a) The $^rQ_4$ branch of DSSD. The stepwidth is 0.08 MHz. (b) Zoom display of the bandhead.

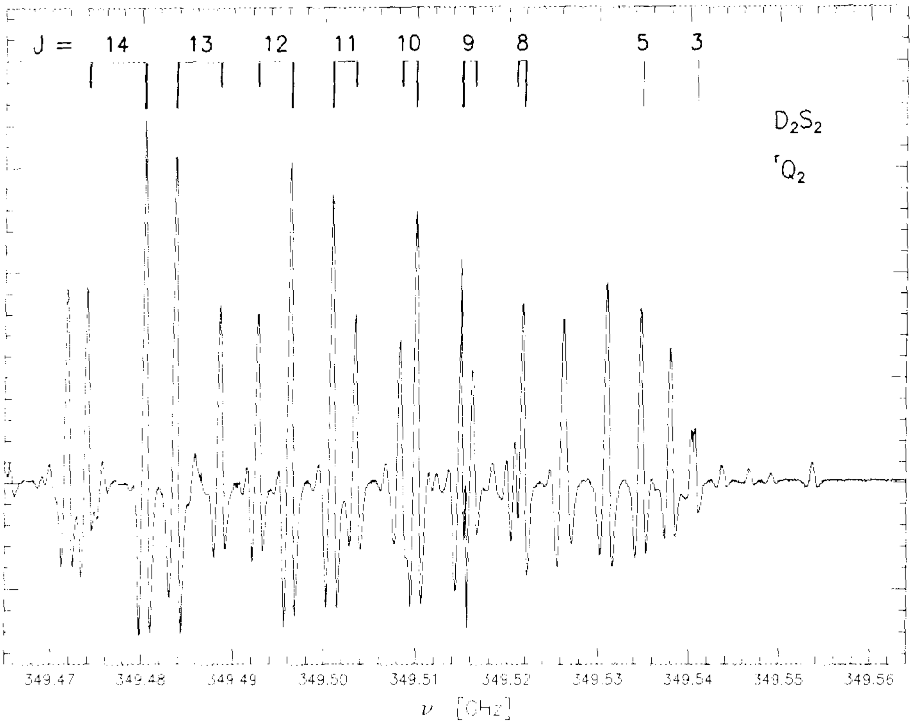

FIG. 4. The bandhead of the $^rQ_2$ branch of DSSD displays the K-doubling which causes its complex appearance. In fact the observed splitting is dominated by the centrifugal distortion splitting and is anomalous. This figure should be compared with Fig. 6 of Ref. (7).

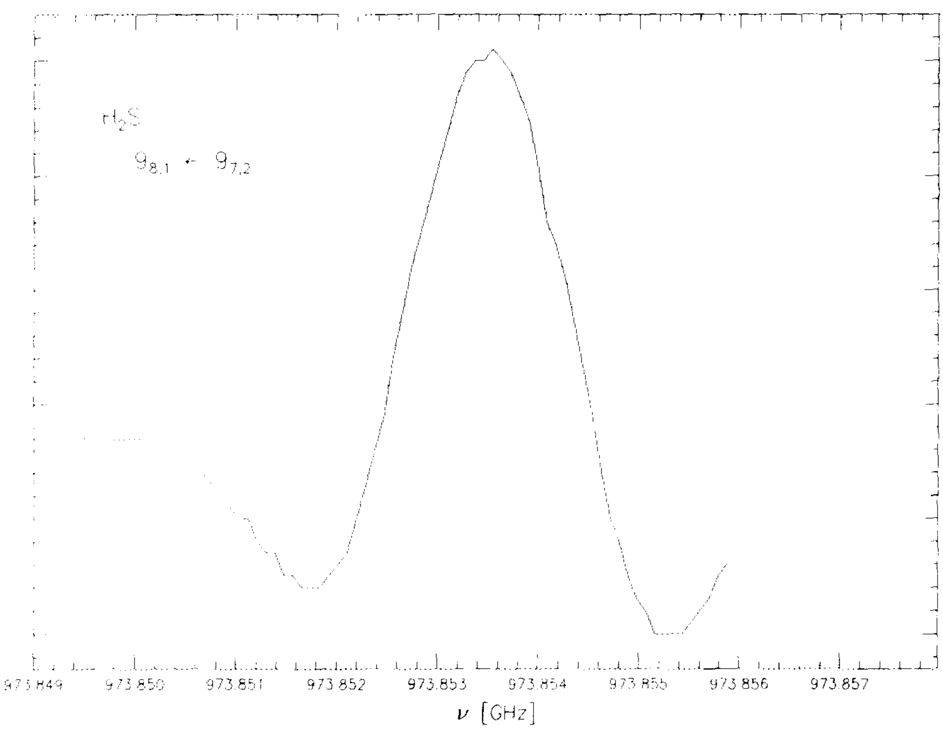

FIG. 5. H_2S transition near 1 THz ($9_{8,1}$–$9_{7,2}$, 973 853.486 MHz).

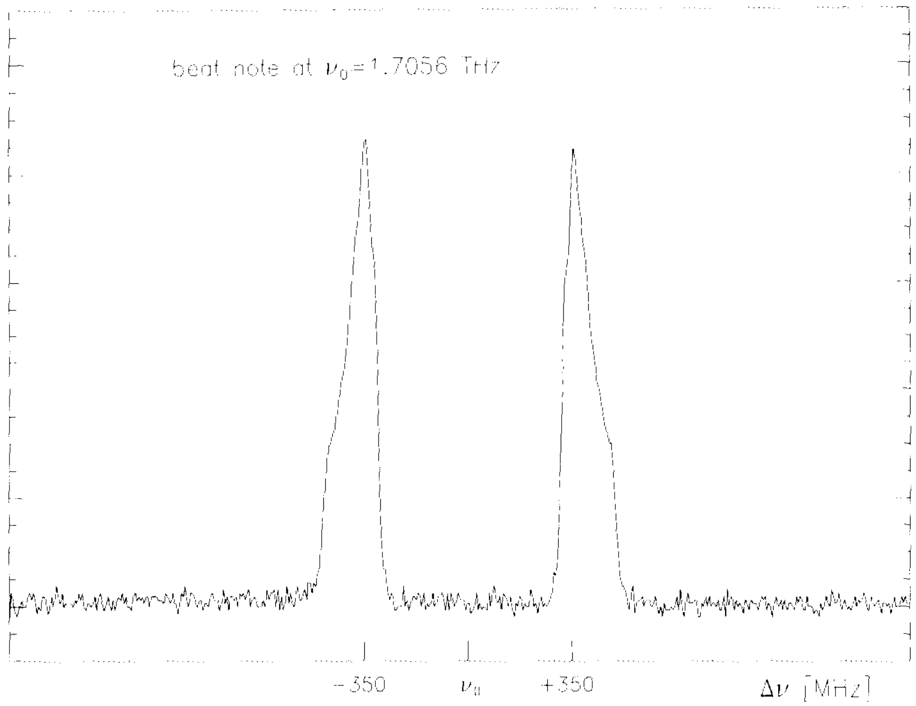

FIG. 6. Beat signal at 1.7056 THz.

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