New Techniques for Submillimeter Precision Broadband Spectroscopy

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We report precision broadband spectroscopic measurements in the spectral region up to 1.1 THz. Continuous frequency coverage is achieved by employing frequency and phase stabilized Backward Wave Oscillators (BWOs). This breakthrough in high-resolution scanning spectroscopy in the terahertz region with microwave accuracy and hitherto unparalled sensitivity became possible by the opening of the borders between East and West and essentially by the collaborative technical efforts between the University of Cologne and Giessen, Germany, and the Institute of Applied Physics, Nizhnii Novgorod, Russia.

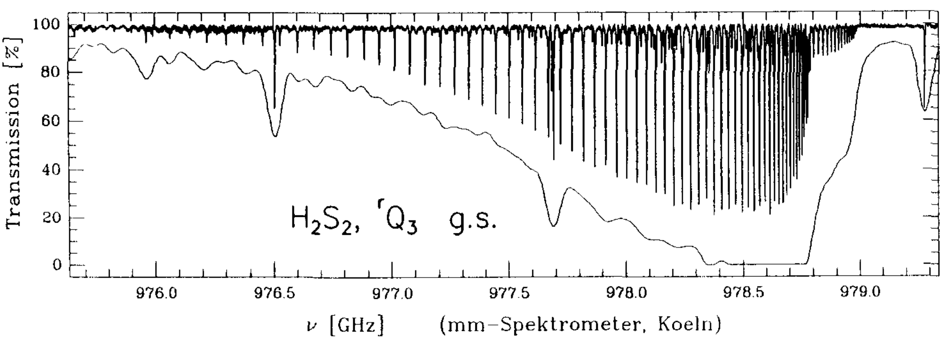

Figure 1: The ${}^{r}Q_3$ branch of HSSH near 1 THz in comparison with a Fourier transform recording (lower trace) obtained in the Giessen laboratory.

The essential components of the Cologne Terahertz spectrometer system consist of high-frequency, broadband tunable BWOs, supplied by the ISTOK Research and Production Company (Fryazino, Moscow region), a newly designed multiplier-mixer with a low noise HEMT amplifier circuitry, two precision tunable millimeter wave synthesizers covering the frequency region between 78 to 118 GHz and 118-178 GHz. They are commercially available from the Institute of Electronic Measurement, KVARZ (Nizhnii Novgorod). A He-cooled InSb-hot electron bolometer is used as a detector.

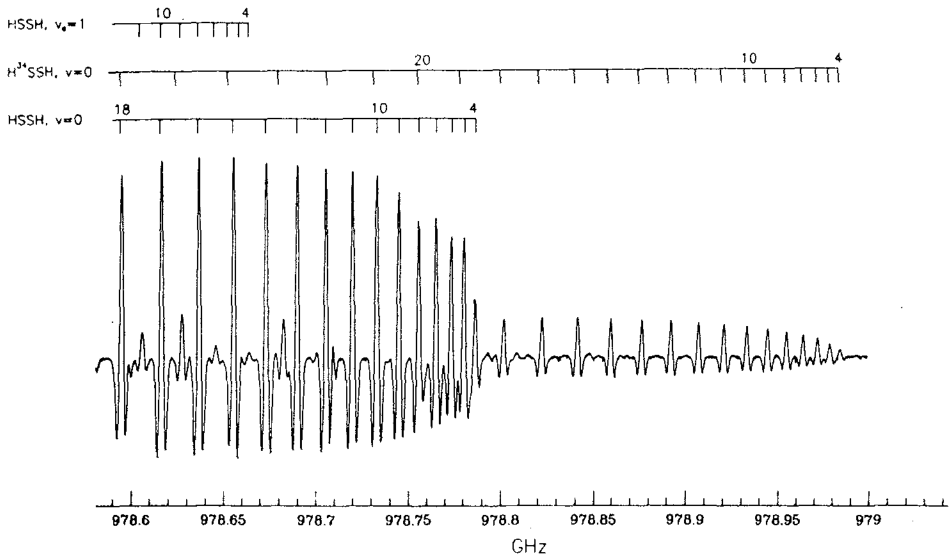

Figure 2: Band head of the ${}^{r}Q_3$ branch of HSSH recorded with a phase locked BWO.

Extensive measurements have been performed on HSSH, HOOH, on their various isotopomers, and on a large number of astrophysically relevant molecules, such as SO_2, H_2S, HCN, H_2CO, HNCS, HNCO, CH_3OH, and radicals SO, CN, NO, and CCH. The pure rotational spectra of simple chain molecules such as HSSH in the ground and various vibrationally excited states are particularly amenable to broadband high-resolution spectroscopy, both in the Doppler limited and in the sub-Doppler domain. The beauty of perpendicular spectra exhibited by near prolate tops and the wealth of information contained in the spectra is revealed impressively by this new technique. We have performed comprehensive measurements of the ${}^{r}Q_{Ka}$ branches for higher $K \geq 2$ values of HSSH and its various isotopomers. In Fig.1 we display the ${}^{r}Q_3$ branch of HSSH, recorded with a free-running THz BWO as radiation source. The trace underneath represents the best recording of this Q branch by high resolution Fourier transform spectroscopy. The band head of this $K_a = 3$ Q branch, registered with a frequency and phase stabilized BWO, demonstrates the high sensitivity of the spectrometer (estimated to be about 10^{-5} cm^{-1}). The appropriate J-assignments are given.

An essential part in the phase-lock-loop (PLL)-system of the Cologne Terahertz spectrometer are the new, continuously tunable frequency synthesizers, which employ a PLL-stabilized BWO, which can be locked to the output of an atomic clock. In the Giessen Microwave Laboratory, the 78 to 118 GHz unit has been employed to record the OCCCS spectrum continuously over the entire tuning range. These automated measurements yielded more than 4000 spectral lines, belonging to the ground and various vibrationally excited states, particularly the excited bending states, the lowest of which is ν_7 at 78 cm^{-1}. The extreme frequency stability of the KVARZ- frequency synthesizer allowed reference spectra to be taken hours later with the cell empty. Subtraction of the two scans yielded the OCCCS spectrum with greatly reduced baselines as shown in Fig 3. Fig. 4 presents a portion of the ${}^{r}Q_0$ branch of HSSH recorded in Cologne with the 118 to 178 GHz KVARZ svnthesizer. With the Cologne system saturation dip spectra have also been obtained.

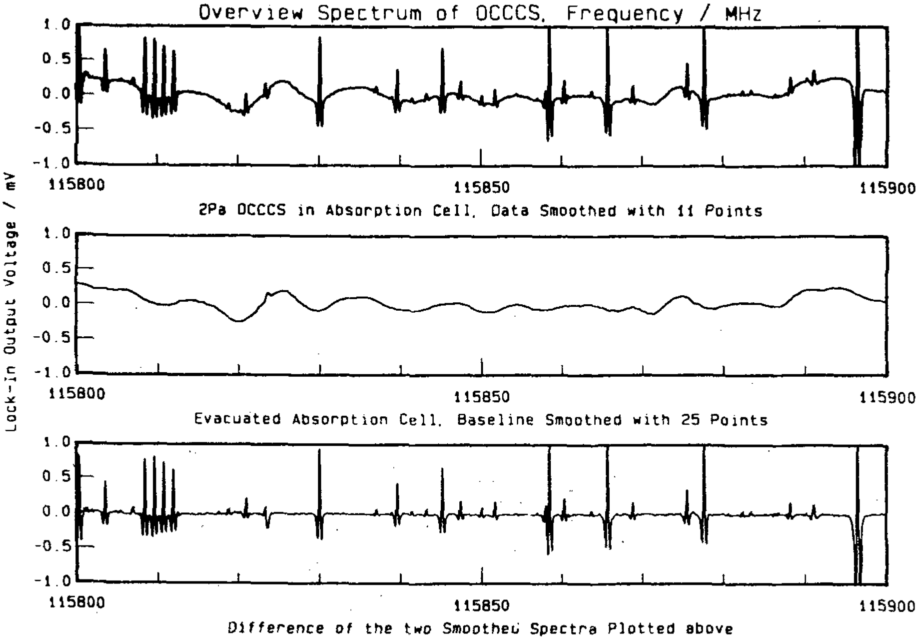

Figure 3: Small part of the frequency scan of OCCCS. Upper trace: filled cell; middle trace: reference spectrum taken with an empty cell; lower trace: difference (filled-evacuated) spectrum. The sensitivity is about 10^{-7} cm^{-1} with a time constant of 120 msec.

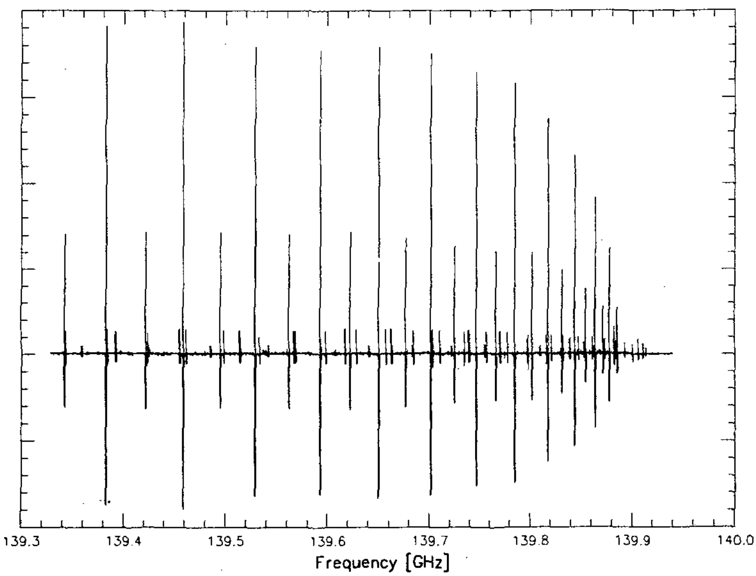

Figure 4: Band head of the ${}^{r}Q_0$ branch of HSSH, recorded with a KVARZ synthesizer. The 3:1 intensity alternation of adjacent J lines is clearly seen.