

A Microwave Fourier Transform Spectrometer in the Frequency Band From 26 to 40 GHz

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We report the design and performance of a microwave Fourier transform spectrometer in the frequency region between 26 and 40 GHz for the investigation of rotational spectra. The performance is illustrated by rotational transitions of allene and allene- d_4 molecules with a very small dipole moment in vibrational excited states. The sensitivity and Doppler limited linewidth is demonstrated by measurements of transitions of isotopomers of carbonyl sulfide and sulfur dioxide.

Introduction

As the result of our experiences with a preliminary spectrometer [1], we have constructed and tested a microwave Fourier transform (MWFT) spectrometer in the frequency region 26 to 40 GHz (V or R band). Although it is more difficult and expensive to build such a spectrometer in V band than in lower frequency bands because of the availability of certain microwave components, we have found it useful to extend the technique because it has been proved by work in both Kiel [2] and Zürich [3] that MWFT spectroscopy has in general higher resolution and sensitivity than Stark spectroscopy [4, 5]. It is especially suited for molecules with small dipole moments. Our experience with MWFT spectrometers in lower frequency bands has been useful in designing and constructing the new spectrometer.

Experimental Set Up

Figure 1 shows the principal set up of the spectrometer. In the ensemble of molecules within the sample cell (5)¹ a macroscopic polarization is created by a short, strong microwave carrier pulse (1–4). The fundamental microwave (MW) pulse is produced at half the frequency needed to polarize the molecules. It is then doubled in frequency (3) to cover the band from

26 to 40 GHz and amplified (4). For the detection of the transient signals emitted by the molecular ensemble a superheterodyne system (6) in the range 26 to 40 GHz is used, followed by a data processing unit (7).

A more detailed description is presented in Figure 2.

A backward wave oscillator (2)² with power supply (1), 12 to 18 GHz or 18 to 26 GHz, is phase stabilized by the synchronizer (4) via a matching network (3). The output of a frequency synthesizer (5), referenced to the system master frequency of 10 MHz, is multiplied in the mixer (9) and mixed with the MW from the signal oscillator (2) via the directional coupler (7). The isolator (6) reduces any modulation of the signal MW. The attenuators (8) and (13) are for power adjustment. The signal MW is checked by the counter (15) combined with a power meter. This part of the spectrometer may be replaced by a microwave synthesizer referenced to the master frequency.

MW pulses with a minimum length of 10 ns are formed by the PIN switches PS 1 and PS 2 (17, 18). The time diagram is given in Fig. 2 of [10]. As they reflect the MW when closed, the isolator (16) reduces interference with the phase stabilization. The 0°/180° phase shift in V band necessary for the phase alternating pulse sequence for the reduction of coherent perturbations [2, 6, 7] is made by a 0°/90° phase shifter in Ku or K band (20). Details are given in Figure 3. In a preliminary version of the spectrometer described in

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¹ Numbers in parentheses refer to Figure 1.

² This, and the following numbers in parentheses refer to Figure 2.

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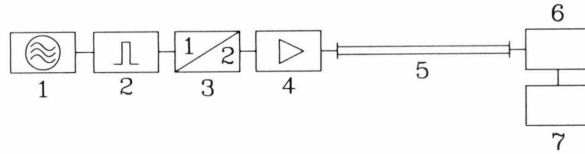


Fig. 1. General set up of a MWFT spectrometer in the range of 26 to 40 GHz.

- | | |
|---|---|
| 1 Phase stabilized MW source from 13 to 20 GHz. | 5 Sample cell. |
| 2 MW pulse generation. | 6 MW superheterodyne detection system with a phase stabilized local oscillator from 26 to 40 GHz. |
| 3 MW frequency doubler. | 7 Data processing system. |
| 4 MW pulse amplification. | |

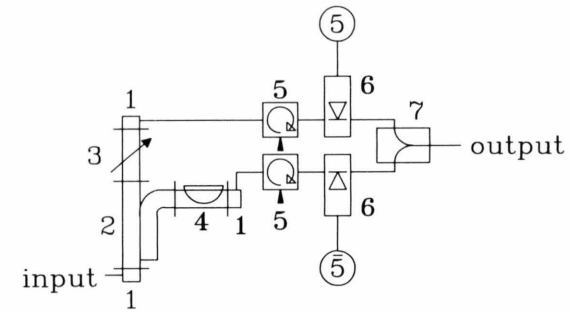


Fig. 3. $0^\circ/90^\circ$ Phase modulator from 13 to 20 GHz.

- | | |
|--|---|
| 1 Coaxial to waveguide transition. | 5 Isolator, Microtek I124K18A2. |
| 2 Directional coupler, Ku band, FXR Y610A. | 6 PIN switches, HP33144A with TTL driver |
| 3 Phase shifter. | 7 Power divider/combiner, TRM DMS 285-20, 1-20 GHz. |
| 4 Waveguide variable attenuator. | |

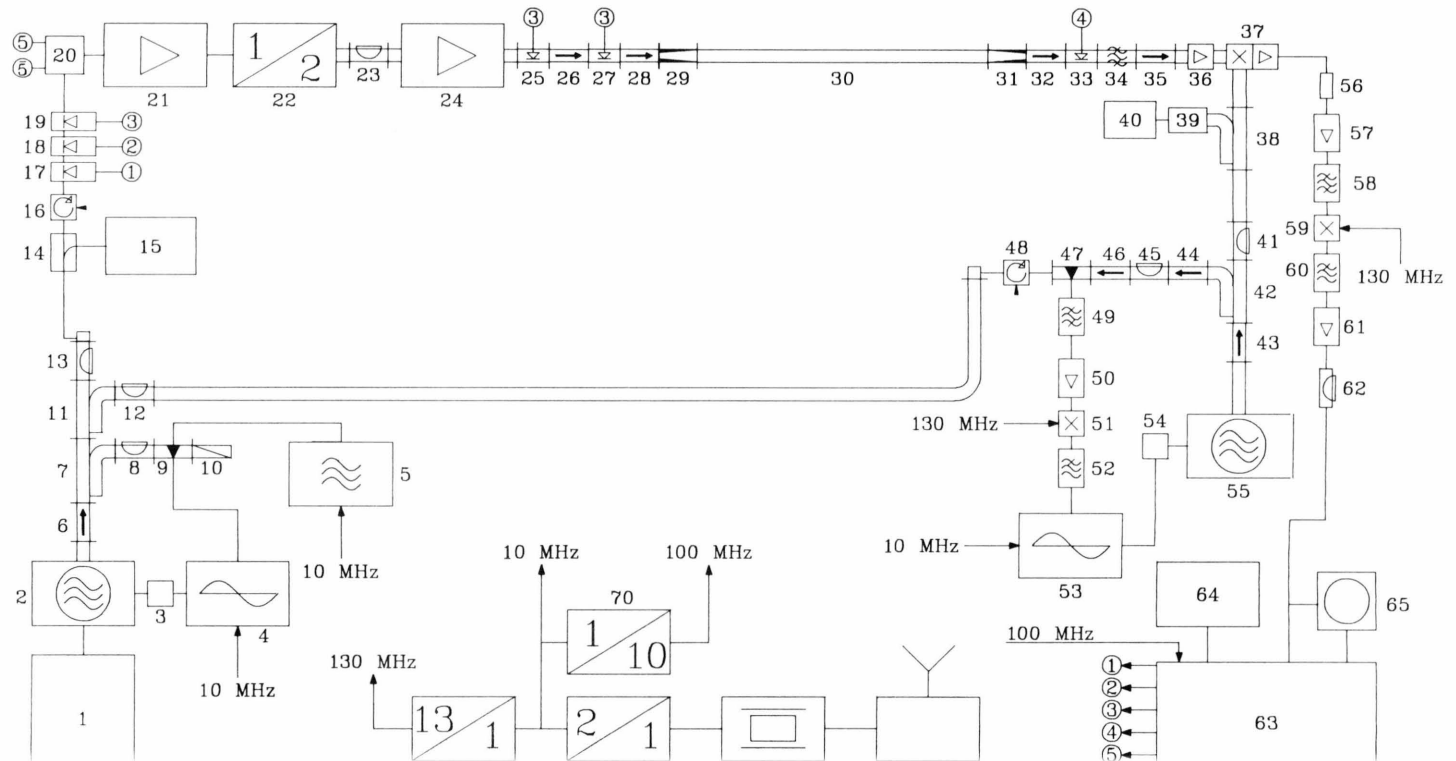


Fig. 2. Detailed set up of a MWFT spectrometer from 26 to 40 GHz. BW: Bandwidth; IF: Intermediate frequency; V: V Band, 26.4–40 GHz; Ku: Ku Band, 12.4–18 GHz; K: K Band, 18–26.4 GHz.

- 1 BWO power supply, Heinzinger; HNS 4000-01, HNCS 600-10, TNS 10-30, TN 100-20, TNS 200-20.
- 2 Signal source, BWO 12.4–18 GHz, Varian 162 Y, or BWO 18–26.4 GHz, Watkins Johnson (WJ) 2057–50.
- 3 Matching network.
- 4 Synchronizer, Schomandl FDS 30.
- 5 Frequency synthesizer, 5–2700 MHz, Rohde & Schwarz (R & S), SMPD.
- 6 Waveguide isolator, Ku.
- 7 Waveguide directional coupler, Ku, 10 dB.
- 8 Variable attenuator, Ku.
- 9 Waveguide mixer with diode 1N26, Ku.
- 10 Waveguide termination, Ku.
- 11 Waveguide directional coupler, Ku, 10 dB.
- 12 Variable attenuator, Ku, Hewlett Packard (HP) P375A.
- 13 Variable attenuator, Ku, HP P375A.
- 14 Coaxial directional coupler, 10 dB, MAC Technology (MAC) C3207-10.
- 15 Frequency counter with power meter, Systron Donner 6030 Parts 1 to 10 and 14, 15 may be replaced by a microwave synthesizer 10 MHz–26.5 GHz HP 8340B.
- 16 Coaxial isolator, SMT C1118.
- 17 PIN switch with TTL-driver, HP 33144A and equivalent to HP 33190B usable to 20 GHz.
- 18 see 17.
- 19 see 17.
- 20 0°/90° phase modulator, see Figure 3.
- 21 Microwave amplifier, HP 8349B.
- 22 Microwave frequency doubler, HP 83554A.
- 23 Variable attenuator, V, Narda V727.
- 24 Travelling wave tube amplifier, 26.4–40 GHz, with incorporated output isolator, Hughes 8010H, present version 5 watt.
- 25 PIN switch, V, AEG PS28 HIS, insertion loss < 1.5 dB, isolation > 65 dB.
- 26 Waveguide isolator, V, Trak 2071-1810, isolation 23 dB minimum, VSWR < 1.15.
- 27 PIN switch, V, AEG PS28 HHI, insertion loss < 1.3 dB, isolation > 70 dB.
- 28 Waveguide isolator, V, see 26.
- 29 Taper from V waveguide to 8 × 8 mm square waveguide.
- 30 Sample cell, square waveguide, cross section 8 × 8 mm, length 3.2 m, with mode filters, vacuum tight MW shielded windows and vacuum system.
- 31 Taper from V waveguide to 8 × 8 mm square waveguide.
- 32 Waveguide isolator, V, see 26.
- 33 PIN switch, V, see 27.
- 34 Band pass filter, V, HP R362A.
- 35 Waveguide isolator, V, see 26.
- 36 Microwave amplifier, Hughes A 1350 H-3001, 26.4–33 GHz, gain 21 dB, noise Fig. 5 dB, or Hughes A 1355 H-3601, 33–40 GHz, gain 17 dB, noise Fig. 6 dB.
- 37 Biased mixer with preamplifier, V, Spacek PKA-U.
- 38 Directional coupler, V, 10 dB, FXR U610CF.
- 39 Power sensor, V, HP 8486A.
- 40 Power meter, HP 435B.
- 41 Variable attenuator, V, De Mornay Bonardi (DB) D430.
- 42 Directional coupler, V, 10 dB, DBD675-10.
- 43 Waveguide isolator, V, see 26.
- 44 Waveguide isolator, V, see 26.
- 45 Variable attenuator, V, Narda V727.
- 46 Waveguide isolator, V, see 26.
- 47 Waveguide harmonic mixer, Spacek 2M2B-200.
- 48 Coaxial isolator, Ku, Teledyne T-12S44-1M.
- 49 Band pass filter, 160 MHz, BW 10 MHz.
- 50 Amplifier, gain 40 dB, Amplica USL401.
- 51 IF mixer, Mini Circuits ZAD1.
- 52 Low pass filter 110 MHz.
- 53 Synchronizer, Schomandl FDS 30.
- 54 Matching network.
- 55 Local oscillator, sweeper, V, HP 8690B + HP 8697A.
- 56 Attenuator, 3 dB, Suhner.
- 57 IF amplifier, Spacek U 160-30.
- 58 Band pass filter, 160 MHz, BW 20 MHz.
- 59 IF mixer, Mini Circuits ZAD1WH.
- 60 Low pass filter 55 MHz.
- 61 Amplifier, GPD 461, 462, 464.
- 62 Variable attenuator.
- 63 Analog to digital converter, averager, and experimental control unit, 10 to 100 ns sampling intervals, 1024 to 4096 data points, 75 kHz repetition rate maximum [12].
- 64 Personal computer, Tandon PCA 20.
- 65 Oscilloscope, R & S, 100 MHz.
- 66 Normal frequency receiver, R & S XKE2.
- 67 Quartz frequency standard, 5 MHz, R & S XSD2.
- 68 Frequency doubler.
- 69 Frequency multiplier.
- 70 Frequency multiplier.

[1] a $0^\circ/180^\circ$ biphas modulator in Ku band was erroneously used. The MW pulses are amplified (21) and fed into a frequency doubler (22). The frequency doubling converts the $0^\circ/90^\circ$ phase shift into a $0^\circ/180^\circ$ phase shift. The travelling wave tube amplifier (TWTA) (24) increases the power of the pulses to a sufficient level to create a strong molecular polarization. This is adjustable with attenuators (13) and (23). The PIN switches PS 3 (19, 25, 27) isolated by the isolator (26) are open during pulse creation and are closed during the detection of the molecular signal to reduce any feed through of CW microwave power from the signal source (2) to the detection system. The pulses create a macroscopic polarization of the molecules in the sample cell (30) of square cross section (8×8 mm) and a length of 3.2 m. Mode filters at both ends of the cell suppress the unwanted degenerate mode polarized perpendicular to the desired one. They are made of metallized mica strips of 10 cm length, arranged parallel to the electric vector of the unwanted mode. The cell is enclosed by MW shielded vacuum tight windows, tapers (29, 31) to normal V band waveguide, and waveguide isolators (28, 32) with low VSWR to reduce pulse reflections. The vacuum system is connected to the sample cell by slits of 0.5 mm width and 10 cm length in the waveguide wall perpendicular to the electric vector of the desired mode. The detection system is protected from the strong MW pulses by the PIN switch PS 4 (33). A waveguide bandpass filter (34) reduces its switch transients. The low noise microwave amplifier (36) amplifies the transient molecular signal. To cover V band, two amplifiers are used, one from 26 to 33 GHz, gain 21 dB, noise figure 5 dB, and the other from 33 to 40 GHz, gain 17 dB, noise figure 6 dB. They are the main contributors to the noise figure of the detection system. The amplified molecular signal is mixed with the microwave signal from the local oscillator (55) in the mixer (37) and is thereby downconverted to a band around 160 MHz.

It is necessary to phase stabilize the local oscillator (55) ranging from 26 to 40 GHz against the signal oscillator (1). As is shown in [1, 11] it is also possible to make the primary stabilization at the local oscillator and to slave the signal oscillator to it. Although this eases the search for sources of coherent perturbations, we prefer to make the primary stabilization at the oscillator with the lower MW frequency. The phase stabilization is made by a second harmonic mixer (47) fed with the signal MW (13 to 20 GHz) via

the directional coupler (11), the attenuator (12) and the isolator (48), and at the same time fed with the local MW (26 to 40 GHz) via the directional coupler (42), the isolators (44, 46) and the attenuator (45). There should be enough isolation to prevent the second harmonic of the signal MW from reaching the detection system via the directional coupler (42). The intermediate frequency of the mixer (47) is band limited to 160 MHz by a bandpass filter (49), amplified (50) and downconverted to 30 MHz by the mixer (51), which is supplied with 130 MHz referenced to the system master frequency. The local MW power can be adjusted with the attenuator (41). It is monitored with the power meter (39, 40) via the directional coupler (38).

The molecular signal, downconverted to a band around 160 MHz by the mixer (37), is amplified (57) and band limited to 160 ± 10 MHz (58). The second downconversion by the mixer (59) converts it to a band of 30 ± 10 MHz, as the signal averager (63) has a Nyquist frequency of 50 MHz. The lowpass filter (60) limits the signal to the range below 50 MHz. Its power is adjusted to the proper input level of the averager by the amplifier (61) (in which is incorporated a 5 MHz highpass filter) and by the attenuator (62). Details of the averager are given in [12]. The averager provides in addition the control sequence for the PIN switches. A personal computer (64) is used to control the experiment, as well as to carry out data handling and Fourier transformation. All frequencies of the spectrometer are referenced to the normal frequency and time station DCF 77 Mainflingen by the receiver (66), the regulated quartz oscillator (67) and multipliers (68, 69, 70).

Figure 3 shows the $0^\circ/90^\circ$ phase modulator working from 13 to 20 GHz. The PIN switches (6)³ open one or the other channel. The isolators (5) suppress reflections at the closed switches. Since the phase shift of the waveguide directional coupler (2) is not exactly 90° , it must be adjusted using the phase shifter (3). The attenuator (4) is necessary to adjust the amplitude balance. Because the polarizing MW pulse can be seen on the oscilloscope ((65) in Fig. 2), the control and adjustment of the amplitude balance and phase shift is possible. The range from 26 to 40 GHz is covered by frequency doubling.

³ This, and the following numbers in parentheses refer to Figure 3.

It would be useful to have a precise and frequency independent $0^\circ/90^\circ$ modulator over the whole range from 13 to 20 GHz or a $0^\circ/180^\circ$ modulator from 26 to 40 GHz.

Experimental Tests and Results

The performance of the spectrometer was tested with allene, allene- d_4 and several isotopic species of carbonyl sulfide in natural abundance.

Vogelsanger recorded rotational spectra of allene and allene- d_4 in various degenerate vibrational states [13]. Only the $J_K \rightarrow J_{K'} = 1_0 \rightarrow 0_0$ rotational transition of each vibrational state was in the range of his MWFT spectrometer. To confirm his assignments, he carried out MW–MW double resonance experiments. The $J_K \rightarrow J_{K'} = 2_0 \rightarrow 1_0$ transitions were used as pump transitions. Their frequencies could be determined by slightly varying the pump frequency and observing the resulting spectrum. Details are given in [13].

We confirmed Vogelsanger's assignments by direct measurements of the rotational transitions $2_0 \rightarrow 1_0$ of allene and allene- d_4 in the vibrational states $v_{11}=1$ and $v_{10}=1$. The results are given in Table 1. They show that observing spectra of molecules with very small dipole moments is feasible with our spectrometer. One example of a spectrum is given in Figure 4. It shows the $2_0 \rightarrow 1_0$ transition of allene- d_4 in the vibrational state $v_{11}=1$. The dipole moment of the molecule in this state is only 0.005(2) Debye [13].

For direct comparison with our earlier V band spectrometer [1] we remeasured the $J \rightarrow J' = 3 \rightarrow 2$ transition of $OC^{33}S$, which has ^{33}S hyperfine structure. We needed 10 k averaging cycles to reach a good signal to noise ratio. The spectra are shown in Figure 5. No coherent perturbations are visible. As can be seen from Fig. 3 of [1], 6000 to 30 000 k averaging cycles were necessary with the preliminary set up to reach a much poorer signal to noise ratio with perturbations dominating some of the spectra. This is clear proof of the great performance enhancement of our new spectrometer. The $J \rightarrow J' = 3 \rightarrow 2$ transition of ^{18}OCS in the vibrational state $v_2=2$ with an absorption coefficient of $7.3 \cdot 10^{-10} \text{ cm}^{-1}$ at a temperature of -50°C was detected with 1500 k averaging cycles.

The achievable linewidth was another point of interest. We measured the $4_{04} - 3_{13}$ transition of sulfur dioxide at a pressure lower than 0.1 mTorr (0.014 Pa).

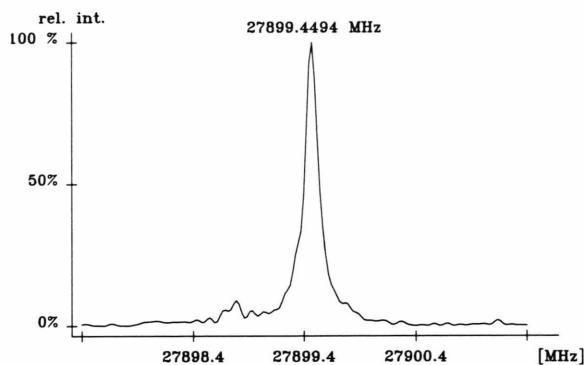


Fig. 4. A 4 MHz section out of a 50 MHz scan of the rotational power spectrum of the $J_K \rightarrow J_{K'} = 2_0 \rightarrow 1_0$ transition of allene- d_4 in the vibrational state $v_{11}=1$. Sample interval 10 ns, 2600 k averaging cycles, pressure 7 mTorr (0.9 Pa), temperature -44°C , polarization frequency 27 899.4 MHz, 1024 data points supplemented with 3072 zeros prior to Fourier transformation.

Table 1. Observed transition frequencies [MHz] of allene and allene- d_4 .

Molecule	Vib. state	Transition $J_K \rightarrow J_{K'}$	Our results	Vogelsangers results [13]
Allene	$v_{11}=1$	$2_0 \rightarrow 1_0$	35 617.623(1)	35 617.620(10)
	$v_{10}=1$	$2_0 \rightarrow 1_0$	35 517.203(1)	35 517.190(10)
Allene- d_4	$v_{11}=1$	$2_0 \rightarrow 1_0$	27 899.449(2)	27 899.44 (10)
	$v_{10}=1$	$2_0 \rightarrow 1_0$	27 843.781(2)	27 843.780(30)

Pressure broadening is negligible at this low pressure. The transition has a half width of 24 kHz at half height in the power spectrum (see Figure 6). This experimental result is in very good agreement with the calculated half width using Mäder's theory [14]. According to his article, Doppler broadening is the dominant linewidth factor, but there is also a contribution from wall collision broadening. Because both broadening factors make increased contributions at higher frequencies, the resolution capability of the spectrometer is not so good as in lower frequency bands. Further measurements have been made with iodoacetylene, perdeutero-methylamine, 1,2-dichlorobenzene, isopropylamine, hydrazoic acid, and many other molecules. The results of these measurements are to be published.

Summarizing our results we can state that our present set up of a V band spectrometer has reached the level of performance we normally obtain from our lower band MWFT spectrometers. The only exception is the poorer resolution of narrow multiplets. The spectrometer is especially suited for the recording of spectra of molecules with very small dipole moments.

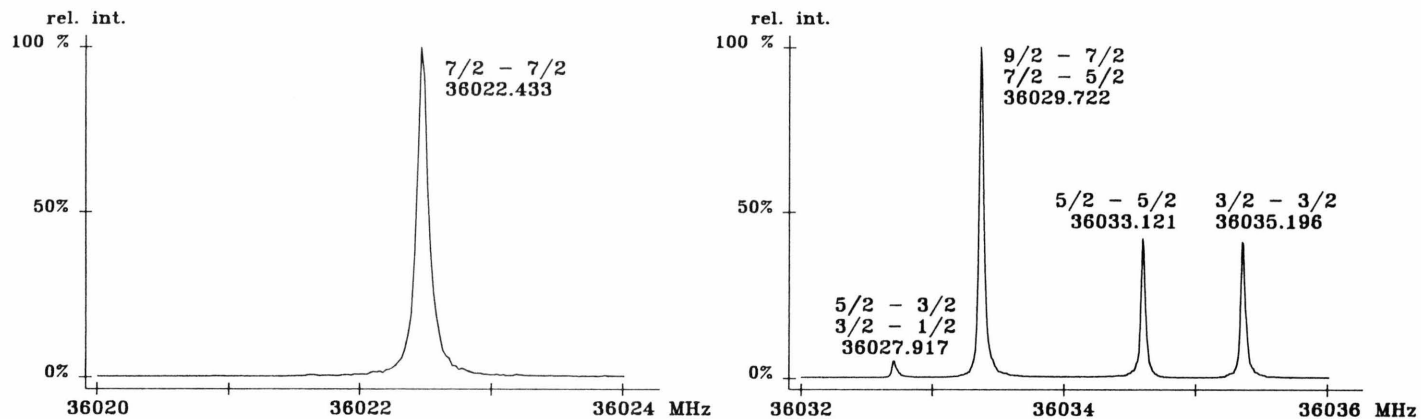


Fig. 5. Hyperfine structure of the rotational transition $J \rightarrow J' = 3 \rightarrow 2$ of ^{33}S -carbonylsulfide in natural abundance. The lines were polarized with two different polarization frequencies, 36 022 MHz (left) and 36 036 MHz (right). Sample interval 10 ns, 10 k averaging cycles, pressure 6 mTorr (0.8 Pa), temperature -53°C , 2048 data points of the transient decay supplemented with 2048 zeros prior to Fourier transformation.

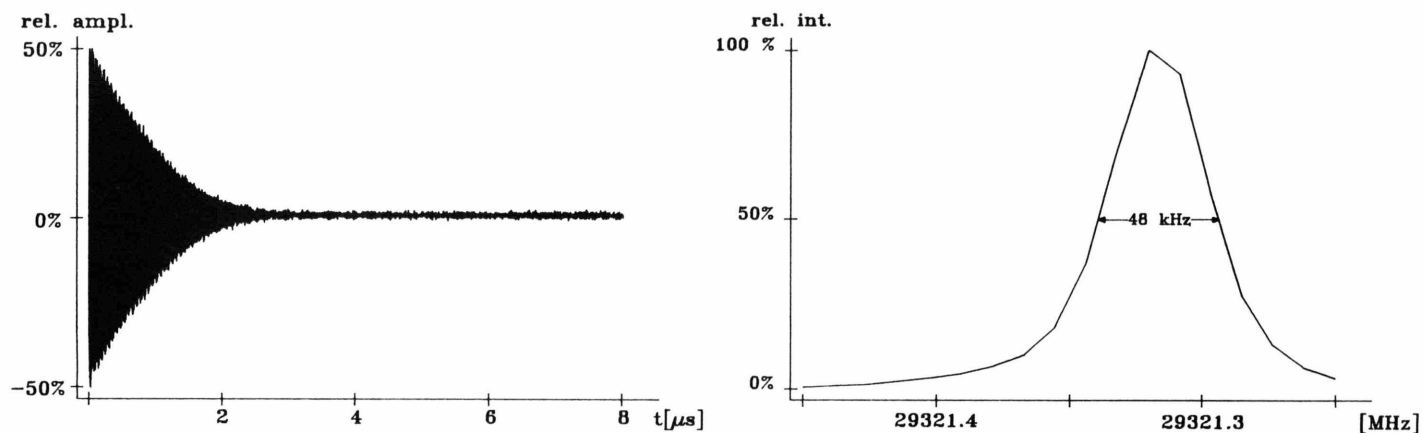


Fig. 6. Time domain signal (left) and power spectrum (right) of the rotational transition $4_{04} - 3_{13}$ of sulfur dioxide in the vibrational ground state. Sample interval 20 ns, 30 k averaging cycles, pressure < 0.1 mTorr (0.014 Pa), temperature -50°C , polarization frequency 29 321 MHz, 4096 data points recorded. The time scale must be multiplied by 10.

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