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January 1986

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PULSED MILLIMETER WAVE FOURIER TRANSFORM MICROWAVE
SPECTROMETER

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ABSTRACT

An improved pulsed microwave spectrometer for the detection of rotational transitions in gaseous molecules in the frequency range of 130-150 GHz is described. It incorporates a tunable Fabry-Perot cavity and a low noise superheterodyne receiver for the detection of the molecular emission signals. The molecules are excited by $\pi/2$ pulses provided by a high efficiency frequency doubler which is pulse modulated at an IF frequency of 1.4 GHz.

INTRODUCTION

In pulsed Fourier transform spectroscopy¹ a brief intense pulse of microwave energy is used to polarize the molecules over a band of frequencies. These molecules then emit a polarization decay signal which is detected by a sensitive receiver. After signal averaging, the absorption spectrum can be obtained by Fourier transformation. The method is directly analogous to that used in pulsed nuclear magnetic resonance (NMR) experiments, and has many of the advantages associated with that technique.

In recent years, a number of pulsed spectrometer systems^{1,2,3} have been developed. These have been limited to operating frequencies below 18 GHz because of the unavailability of suitable switches (on-off ratio greater than 100 dB and rise time of a few ns.) to form the microwave pulse. In the present spectrometer, instead of switching the microwave power on and off directly, a high efficiency frequency doubler⁴ is pulse modulated using a conventional low frequency switch to produce sidebands with a very high on-off ratio. A high Q cavity containing the gas is then tuned to one of the sidebands to produce the desired polarization.

A preliminary version of this spectrometer has already been described⁵. In the present paper, a number of improvements to the system are discussed. These include the use of a lower noise receiver with an IF frequency of 1.4 GHz, a signal digitizer with very fast averaging capabilities and a track-and-hold feedback system to stabilize the cavity frequency during the brief excitation pulses. These improvements lead to enhancements in performance which are illustrated below.

SPECTROMETER DESCRIPTION

A block diagram of the improved spectrometer is shown in Fig. 1. The major components used include a 70-GHz microwave source, a modulated frequency doubler providing excitation pulses at 140 GHz, a low-noise receiver to detect the molecular emission signals, and a Fabry-Perot transmission cavity containing the sample gas.

A klystron, phase locked to a frequency synthesizer, provides microwave power over the frequency range 67-73 GHz. A fraction (10%) of the klystron output serves to drive a frequency doubler providing local oscillator power for the 140 GHz receiver. The remaining source power, up to 80 mW, is used to generate the molecular excitation pulses.

To provide sufficient pulse power to polarize the gas molecules, a tunable high efficiency frequency doubler constructed according to a design⁴ furnished by the National Radio Astronomy Observatory is employed. This doubler has a conversion efficiency of at least 15% over the frequency range of interest, yielding up to 12 mW at 140 GHz. The doubler is modulated at 1.4 GHz by capacitively coupling the modulation source to the doubler bias port.

With a modulation input power of 10 dBm and appropriate adjustment of the DC bias, up to 3 mW can be obtained in each of the sidebands above and below the 140 GHz doubled frequency.

The 1.4 GHz modulation signal is produced using the arrangement shown in Fig. 2. The output power of a crystal controlled oscillator at 1.37 GHz is divided into two equal parts. One part is reserved as a 1.37 GHz source for the receiver IF system as described below. The remainder is upconverted to 1.4 GHz by mixing it with 30 MHz derived from the frequency synthesizer master oscillator. A filter tuned to 1.4 GHz with a bandwidth of 20 MHz removes the unwanted frequency components. After amplification, the signal is attenuated to provide the desired modulation power. The 1.4 GHz reference signal is pulsed on and off using a Watkins-Johnson model S1 switch to control the 30 MHz input to the mixer. This switch has an on/off ratio greater than 90 dB and a rise time of 1 ns when operated at 30 MHz.

The 140 GHz carrier and both modulation sidebands are fed to a tunable Fabry-Perot transmission cavity containing the molecular species to be detected. The construction of the cavity is described in detail in Refs. 5 and 6. It consists of two opposing mirrors 5 cm in diameter, one flat and the other spherical, mounted in a semi-confocal configuration with a distance of about 7.4 cm between the two mirrors. Two coupling waveguides are attached to the flat mirror through electroformed coupling holes. The cavity can be tuned by moving the spherical mirror with the use of a piezoelectric transducer and a stepping motor fitted with a micrometer drive. The loaded Q of the cavity is about 73000. Because of the high quality factor only one sideband from the modulated frequency doubler passes through the cavity and reaches the receiver. By appropriately tuning the cavity either sideband can be detected.

After passing through the cavity the microwave signal is converted to an IF frequency of 1.4 GHz by the receiver. A commercial mixer, Millitech model MXW-6F-1401, with a SSB noise figure of about 8 dB is used. Local oscillator power for the mixer is provided by a second, unmodulated frequency doubler as described above. After preamplification, the IF signal is downconverted to 30 MHz by mixing it with the 1.37 GHz reference. A 1.4 GHz filter removes receiver IF noise contributions at 1.34 GHz.

A portion of the 30 MHz IF signal is used to control the cavity tuning. This is accomplished by phase detecting the IF signal with a mixer driven by the 30 MHz reference source. The phase of the reference is chosen to detect the quadrature component of the cavity transmission signal. The quadrature signal is proportional to the deviation of the cavity frequency from resonance. Since the spectrometer is pulsed, the signal is sampled during the pulse and held constant between pulses using a commercial track-and-hold circuit (Analog Devices model HTC-0300). It is then fed to the piezoelectric transducer to control the cavity frequency. An appropriate filter/amplifier maintains the feedback loop stability.

The remaining portion of the IF signal is further amplified to detect the weak transient molecular emission. To protect the IF amplifier from overload during the much stronger excitation pulse, a gating switch with a 35 dB on/off ratio is employed. It is turned on a few hundred ns after the end of the pulse. The amplified signal is then phase detected using the 30 MHz reference with its phase adjusted to obtain the in-phase component of the cavity signal. Following the phase detector, a low-pass amplifier with a bandwidth of 5 MHz removes all the remaining high frequency components of the signal. The signal is then converted to digital form using an 8-bit transient

digitizer. The digitizer, Transiac model 2001, is a CAMAC based module with a sampling rate of up to 100 MHz and a memory size of 8K channels. The digitized signals from a number of sweeps are then averaged to obtain an adequate signal-to-noise ratio. To minimize the time between pulses, a hardware averager, Transiac model 4100 is used. This averager can accumulate up to 64000 sweeps with an overhead of about 400 ns per sampled point, about 75 times faster than can be accomplished using software averaging techniques. After signal averaging, the accumulated data is transferred to the computer (PDP 11/34) for display and further processing.

To provide the necessary signals to drive the two switches, control the track-and-hold circuit and trigger the transient digitizer, a pulse sequencer was constructed. It was interfaced to the CAMAC system and incorporated two Advanced Micro Devices Am 9513 timing controller chips after the fashion described in Ref 7. In a typical measurement the following sequence is employed: First the IF amplifier is disabled by the gating switch after which the excitation signal is turned on for about 400 ns. At the peak of the cavity signal, the track-and-hold circuit is enabled for 200 ns and at the end of the excitation pulse, the transient digitizer is triggered. Finally, after the excitation pulse in the cavity decays (about 500 ns after the pulse), the IF amplifier is re-enabled and the transient emission signal is digitized and averaged.

MEASUREMENTS

The generation of transient emission signals by the spectrometer is illustrated in Fig. 3. The cavity was filled with SO_2 gas at a pressure of 7.0 mTorr and the microwave frequency was adjusted to 140.3062 GHz, corresponding

to the 6(2,4)-6(1,5) rotational transition of the gas⁸. The excitation pulse width was set to 500 ns and the modulation power at 1.4 GHz was adjusted to maximize the amplitude of the emission signal. In the upper portion of the figure is shown the resulting cavity response and molecular emission signal measured at the input of the IF amplifier. In this illustration the transient emission signal is large enough not to require the use of the IF gating switch. As can be seen, the excitation pulse, shown at the bottom of the figure, is distorted by the cavity time constant. However, it has been shown¹ that the polarization produced is, to first order, proportional to the integral of the pulse independent of this distortion. In order that the molecular emission signal be observed, the cavity decay time must be short compared to the molecular relaxation time. The voltage decay time is given by $\tau_c = 2Q_L/\omega_0$, where Q_L is the cavity loaded Q and ω_0 is the frequency. For our cavity τ_c is approximately 175 ns.

As discussed in Ref. 1, the molecular polarization produced is proportional to the product of the pulse width, microwave electric field amplitude and the rotational transition matrix element. As in the analogous pulsed NMR case, maximum polarization occurs when this product is approximately equal to $\pi/2$. In principle, it is possible to measure or calculate all of these quantities to determine the optimum conditions for the measurement. In practice it is more convenient to choose an appropriate pulse width and to adjust the modulation power for maximum response.

Figure 4 shows the digitized transient emission signal for the 16(2,14)-16(1,15) transition⁸ of the 4.2% abundant isotope of sulfur dioxide, ³⁴S₂ at 141.653 GHz. The data was obtained at a pressure of 3.2 mTorr by averaging 50,000 pulses of width 400 ns and subtracting the

baseline data with the gas removed. In the figure, the excitation pulse ends at $t_p = 0$ and the IF amplifier is gated on at time, t_1 . In curve (a) the excitation frequency was equal to the resonance frequency of the gas. If the frequency is shifted by 1.0 MHz as shown in (b), the emission signal appears as a damped sine wave of that frequency. In either case, the frequency domain signal can be recovered by Fourier transformation.

The decay of the transient emission signal is given to a good approximation by¹

$$E(t) = E_0 \exp(-t/T_2) \exp(-t^2/4q^2). \quad (1)$$

It results from collisional relaxation of the molecules (T_2) and doppler dephasing of the polarization (q) due to molecular motion within the cavity. The quantity, q can readily be computed from the molecular constants, yielding⁵ for SO_2 at 140 GHz, $q = 1.2 \mu s$. The collisional relaxation time, T_2 , which is inversely proportional to pressure, can be expressed in terms of the low power collision broadened linewidth.

As a final example, Fig. 5 shows the transient emission signals at 143.05 GHz for a dilute mixture (117 ppm) of SO_2 in air measured at three different pressures. Also shown in the figure are the results of fitting Eq. 1 to the data using the value, $q = 1.2 \mu s$ predicted for SO_2 . The fitted parameters E_0 and the reciprocal of the relaxation time, $1/T_2$ are plotted in Fig. 6. As can be seen, good agreement with the expected linearity of these parameters with pressure is found. The relaxation time, T_2 can be expressed¹ in the frequency domain as $\Delta f = 1/(2\pi T_2)$ yielding a collisionally broadened linewidth for SO_2 diluted in air of 4.6 MHz/Torr.

In Fig. 5 the data consists of 50,000 samples collected in 256 channels for a total acquisition time of 5.4 sec. The signal to rms noise ratio of the data is about 20:1. By fitting the measured data to Eq. 1, the species concentration (proportional to E_0) can be determined to somewhat greater precision. In spite of the use of a hardwired averager, a significant improvement in performance could still be obtained by employing a faster averager to reduce the dead time between sweeps. With the present arrangement the time necessary to acquire and average a single sweep is about 100 μ s. With faster averaging, this time could be reduced to 10 μ s while still maintaining the requirement that the molecular system relax to equilibrium between sweeps. With this further improvement it should be possible to measure species concentrations in the parts per billion range.

SUMMARY

We have described an improved pulsed microwave spectrometer for the detection of gaseous molecular species. The sensitivity and stability of the apparatus is considerably better than previously obtained⁵. The use of the apparatus to make accurate measurements of the Doppler and collisional relaxation times has been demonstrated. Further improvements in performance can be obtained by employing a faster averager to reduce the dead time between sweeps.

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REFERENCES

1. E.J. Cambell, L.W. Buxton, T.J. Balle and W.H. Flygare, J. Chem. Phys. 74, 813 (1981).
2. A.C. Legon and L.C. Willoughby, Chem. Phys. 74, 127 (1893).
3. W. Stahl, E. Fliege, H. Dreizler and R. Schwarz, Z. Naturforsch. 39a, 354 (1984).
4. J.W. Archer, IEEE Trans. Microwave Theory Tech. MTT-29, 552 (1981).
5. W.F. Kolbe and B. Leskovar, Rev. Sci. Instrum. 56, 97 (1985).
6. W.F. Kolbe and B. Leskovar, Int. J. Infrared and Millimeter Waves 4, 733 (1983).
7. H. Saint-Jalmes and Y. Barjhoux, Rev. Sci. Instrum. 53, 1 (1982).
8. F.J. Lovas, J. Phys. Chem. Ref. Data 7, 1445 (1978).

Figure 1. Block diagram of spectrometer.

Figure 2. Block diagram of IF reference system.

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Figure 3. Cavity excitation pulse and SO_2 transient emission signal for pulse width of 500 ns and gas pressure 7 mTorr.

Figure 4. Transient emission signals for $^{34}\text{SO}_2$ at (a) zero offset frequency and (b) offset of 1.0 MHz.

Figure 5. Measured and fitted transient emission signals for 117 ppm SO₂ in air at various pressure.

Figure 6. Pressure dependence of fitted signal amplitude and relaxation rate of data from Fig. 5.

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