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An Automated Millimeter-Wave Resonator Spectrometer for Investigating the Small Absorption in Gases

V. N. Shanin, V. V. Dorovskikh, M. Yu. Tretyakov, V. V. Parshin, and A. P. Shkaev

Institute of Applied Physics, Russian Academy of Sciences, ul. Ul'yanova 46, Nizhni Novgorod, 603950 Russia
http://www.appl.sci-nnov.ru/mwl/

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Abstract—A resonator spectrometer developed by the Institute of Applied Physics for broadband measurements of the radiation absorption in gases is described. The acquisition and processing of experimental data, as well as the radiation source of this spectrometer, are controlled by the automation system. The setup is used in real-time measurements of the molecular absorption lines in gases at near-atmospheric pressures, specifically, for the investigation of radiation absorption in the open atmosphere.

The precision broadband (45–200 GHz) resonator spectrometer [1–3] developed by the Institute of Applied Physics of the Russian Academy of Sciences (IAP RAS), opens up new fields of use in microwave spectroscopy and its applications [4]. It has been employed to investigate the oxygen absorption band at ~60 GHz [2], the water line at ~183 GHz [5, 6] and the oxygen line at ~118 GHz [7, 8].

The sensitivity of the spectrometer, which is an order of magnitude higher than the sensitivity of available world analogs, has made it possible to not only determine the parameter of the dry-air broadening of these atmospheric lines with the highest up-to-date accuracy [5–7] but also find and measure the line shifts caused by the pressure of atmospheric gases [6, 7]. In atmospheric investigations, it has become feasible to study not only the absorption lines, but also the nonresonance absorption in the atmospheric windows of transparency [5, 8].

In the future, this will provide a possibility of investigating the dynamics of atmospheric processes in the real-time mode (atmospheric monitoring), discriminating between the effects that are responsible for the absorption of electromagnetic radiation by the atmosphere, and studying the dependence of these processes on environmental conditions. The high speed of measurements, the high sensitivity, and the volumetric character of measurements offer ample scope for using the spectrometer in humidity measurements, including direct measurements of the low humidity in the upper atmosphere from an aircraft and low-humidity monitoring in industrial processes, e.g., the manufacturing of optical fibers, handling of fine materials (flour, cement, and gunpowder), etc. In addition, the spectrometer is used to measure radiation absorption in condensed media [9].

Record parameters of the spectrometer have been achieved, because a synthesizer with a fast digital frequency scanning (developed by the IAP RAS) was used as a radiation source for the first time in a setup of this kind. The fundamental necessity of using a dedicated processor to control the synthesizer frequency and the impossibility of utilizing standard software has demanded that a system for automating the experiment should have been developed on the basis of a personal computer (PC).

1. DESCRIPTION OF THE SETUP

Figure 1 shows a block diagram of the resonator spectrometer for the real-time investigations of the absorption of millimeter-wavelength radiation in the atmosphere.

The sensor of the spectrometer is a Fabry–Perot resonator with a length of 25–42 cm, 12-cm-diameter spherical mirrors, and a 24-cm radius of curvature. A TEM (00*q*) wave (*q* is the longitudinal-wave number, i.e., the number of half-waves between the mirrors) is excited in the resonator through a 6-μm-thick Teflon coupling film placed at an angle of 45° with the resonator axis. The Q factor of the resonator is determined by the unavoidable losses due to the reflection from the silver-plated mirrors (silver is the best reflecting material for millimeter wavelengths), and its value is $\sim 6 \times 10^5$.

A backward-wave oscillator (BWO) is used as a radiation source. Three BWOs span the 45–200 GHz band. The BWO frequency is determined by the summed voltage applied from the power supply and the output amplifier of the synchronizer.

The power supply of the BWO is controlled through the high-speed (250 kBaud) RS485 port, which is electrically isolated by a photon-coupled pair and has a FIFO buffer of a 64-byte capacity. The RS485 port is a higher-speed analog of the RS232 port; in addition, it allows up to 32 different devices to be connected. Structurally, it is an interface card connected though the

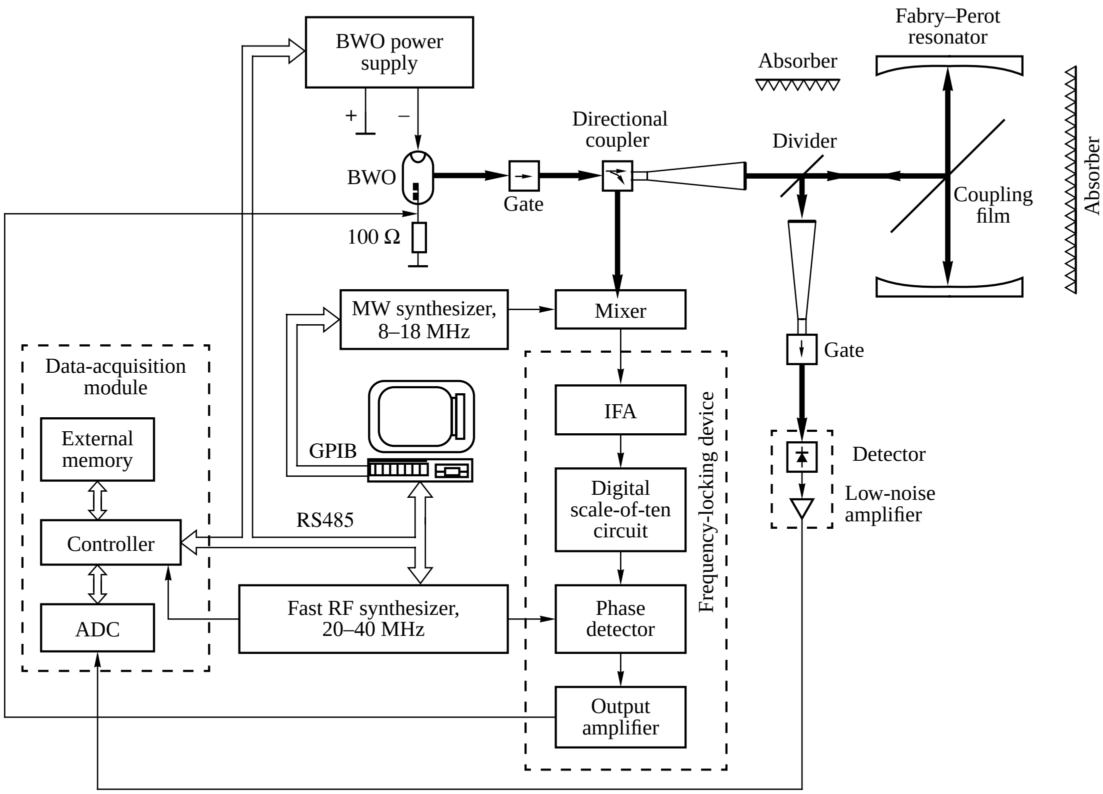

Fig. 1. The block diagram of the wide-range resonator spectrometer.

ISA slot to the motherboard of the PC. The power-supply voltage is set with a step of ~0.2 V; at a steepness of the BWO characteristic of 20–100 MHz/V, this corresponds to a change of 4–20 MHz in the frequency. The uncertainty of the BWO frequency caused by the instability of the power-supply voltage is 10–20 MHz and, sometimes, can reach a value of 100 MHz.

The BWO frequency is stabilized using a phase-lock-in loop (PLL). The BWO radiation phase is locked using the signals of a microwave (MW) synthesizer (8–18 GHz) [2], which determines the center frequency of the BWO, and a radio-frequency (RF) synthesizer (20–40 MHz), which is used for the fast digital scanning of the BWO frequency around the center frequency set by the MW synthesizer. The PLL reduces the uncertainty of the BWO frequency down to ~1 kHz.

The system stabilizing the BWO frequency operates as follows. A portion of the high-frequency power of the BWO at a frequency $f_{\rm BWO}$ is fed to a mixer through a directional coupler. The radiation from the MW synthesizer at a frequency $f_{\rm MW}$ comes to the other input of the mixer. The currents with the relevant frequencies and their harmonic frequencies are induced in the diode of the mixer. The preset frequencies $f_{\rm BWO}$ and $f_{\rm MW}$ are such that the intermediate frequency generated in the diode $f_{\rm if} = nf_{\rm MW} - f_{\rm BWO}$ falls into the passband of the intermediate-frequency amplifier (IFA) with a center frequency $f_{\rm if}^0$ = 350 MHz. The intermediate-frequency signal from the mixer output arrives at the input of the IFA of the FLD.

Upon amplifying, the signal from the IFA output goes into the digital divider by a factor of ten and, then, into a phase detector. The phase difference between the signals arriving from the IFA with frequency $f_{\rm if}/10$ and the signals from the RF synthesizer with frequency $f_{\rm rf}$ is converted in the phase detector into a control voltage, which is applied to the BWO as a feedback signal. As a result, in the case of the operating PLL system, the BWO frequency is

$$f_{\rm BWO} = nf_{\rm MW} - 10f_{\rm rf}, \quad (1)$$

where n is the integer corresponding to the number of the MW-synthesizer harmonic used to stabilize the BWO. It is selected from the range from 4 to 20 depending on the required frequency.

The setup uses a modernized version of the synchronizer described in [10]. Specifically, it has a wider (200 MHz) passband, which is necessary for the fast scanning in the relevant range. The FLD corrects the voltage applied to the BWO within ±10 V, which corresponds to a change of 0.4–2 GHz in the BWO frequency. This means that the PLL helps carry out the fast scanning of the BWO frequency over this range without changing the voltage applied to the BWO from the power supply.

The FLD, which is an important component of the PLL, comprises the IFA, the phase detector, and a control-signal amplifier, as well as automatic tuning devices with an indication of both the frequency locking and the maintenance of the in-sync mode. In the absence of these components, the long-term operation of the PLL would be impossible.

The operating frequency range of an РЧ6-03 MW synthesizer (IEM Kvarts, Nizhni Novgorod) is 8–18 GHz, the minimum frequency step is 20 kHz, and the time of frequency change is 20 ms. The synthesizer is controlled through a standard AT-GPIB/TNT interface card, which is connected to the PC motherboard through the ISA slot. The card is operated with a driver and the function library for the C++ programming language. These functions are used to control the output signal of the MW synthesizer.

The RF synthesizer based on an AD9850 (Analog Device Inc.) microcircuit of direct frequency synthesis provides fast frequency scanning without breaking the oscillation phase during the shift to another frequency (without phase discontinuity). The RF synthesizer is controlled with a special 89C2051 microprocessor, which makes it possible to change the frequency with a constant rate regardless of the workload of the PC. The minimum time between the frequency steps, determined by the capabilities of the microprocessor, is 58 μs; the time of frequency change is 200 ns; the frequency range is 20–40 MHz; and the minimum frequency step is 0.03 Hz.

The signal may be generated under the conditions of frequency scanning by the sawtooth or triangular law. When used, the RF synthesizer can generate sync pulses in two regimes: (a) at the beginning of each scan, which is convenient for the adjustment of the equipment, and (b) for every step in changing the frequency in order to collect the data during the experiment. The RF synthesizer is connected to the PC through the RS485 port.

The resonator-response signal goes to the square-law detector. Its output signal is amplified by the low-noise amplifier and fed into the data-acquisition module. The AT89C51 (Atmel Inc.) controller synchronizes the data-acquisition process with the change in the RF-synthesizer frequency. In addition, it controls the recording of the data acquired by the analog-to-digital converter (ADC) into the external memory and their transmission to the PC. An AD7892 12-bit ADC reads the data under the command generated by the controller each time it receives the sync pulse from the RF synthesizer. These data are transmitted to the external memory with a 32-Kbite capacity, which allows 16 Kwords (i.e., 16 384 readings) to be recorded.

When the built-in memory is filled, the scanning is halted while the data is transferred into the PC storage. The data-acquisition module is connected to the PC via the RS485 port. It takes ≈0.98 s to transfer the data from the external memory filled to capacity.

2. DESCRIPTION OF THE EXPERIMENT

In order to analyze the spectral lines in a gaseous sample at atmospheric pressure, it is necessary to determine the absorption at frequencies of several successive eigenmodes of the resonator.

The absorption in the sample under investigation governs the amplitude and width of the resonance curve of the resonator. The amplitude being measured depends on the power of the incident radiation, which is unstable due to the nonuniformity of the inherent amplitude–frequency characteristic of the BWO, the temperature dependence of the BWO power, the interference effect in the channel, etc. Taking into account a change in the amplitude would require the use of additional equipment and complicate the measuring and data-processing procedures. The width of the resonance curve is free from these drawbacks and can be determined with a higher accuracy during the data processing using frequency-measuring techniques. Therefore, the value of the absorption at the frequency of the resonator's eigenmode is calculated from the width of the resonance.

For the width of the resonance to be measured, the radiation source stabilized by the PLL is tuned to the frequency of the resonator's eigenmode.

A frequency of the MW synthesizer is selected according to Eq. (1) so that the BWO frequency coincides with the desired frequency of the resonator's eigenmode. The eigenfrequencies of the resonator in the BWO range are computed in the program beforehand by a few iterations from the formula

$$f_k = \frac{c}{2L}\left(q + \frac{1}{\pi}\arccos\left(1 - \frac{L}{R}\right) - \frac{c}{4\pi^2 R f_{k-1}}\right),$$

where k is the iteration number, c is the velocity of light in vacuum, q is the integer that corresponds to the eigenmode number, R is the radius of curvature of the mirrors, and L is the mirror separation. In the first iteration, the average operating frequency of the spectrometer is used instead of f_{k-1}. For each mode, the relevant frequency of the MW synthesizer is determined and written in the data file.

The resonance curve of a given resonator mode, from which the width (FWHM) of the resonance line is obtained during the data processing, is recorded using the fast stepwise scanning of the RF-synthesizer frequency around the center frequency of the resonator mode within about ten widths of the resonance curve. For the scanning to be performed, it is necessary to specify the following parameters for the RF synthesizer: the scanning law, the center frequency (Hz), the initial frequency (Hz), the frequency step (Hz), the hold-up time at the scan point, and the triggering mode. If it is necessary to change these parameters, the generation of a frequency is halted.

A major source of the error in determining the width of the resonance is the drift of its center frequency during measurements. This drift is caused by a change in the environmental parameters, which results in an apparent broadening or sharpening of the resonance curve depending on the direction of the frequency drift with respect to the direction of the frequency scanning. For this error to be minimized, the frequency scanning is carried out in the forward and backward directions, and the results are averaged. When the resonator response is recorded, such reversible scans are repeated a given number of times and are averaged during the processing.

All of the control parameters are computed and transferred to the corresponding devices prior to the scanning in order to reduce the duration of the experiment. After the data are acquired, their processing starts. Then, the BWO and the MW synthesizer are tuned to the frequency of the next eigenmode of the resonator.

In order to stabilize the operation of the PLL while changing modes, the voltage of the power supply of the BWO and the frequency of the MW synthesizer are varied simultaneously. The voltage selected is such that, according to the BWO characteristic, the radiation frequency is the closest to the resonator's eigenfrequency.

3. DATA PROCESSING

The processing of the data obtained for each frequency of the eigenmodes is aimed at determining the absorption factor in the sample at that frequency. The total relative losses of the radiation energy during one traversal of the resonator is the sum of the absorption losses in the sample and the losses in the individual resonator components:

$$P_{\text{tot}} = P_{\text{samp}} + P_{\text{refl}} + P_{\text{diff}} + P_{\text{coupl}} = P_{\text{samp}} + P_{\text{res}},$$

where P_{samp} are the absorption losses in the sample, P_{refl} are the reflection losses in the mirrors of the resonator, P_{coupl} are the coupling losses, P_{res} are the resonator losses, and P_{diff} are the diffraction losses due to the escape of radiation from the aperture of the mirrors. As is known, the dependence of the total losses on the width of the resonance curve is defined as

$$P_{\text{tot}} = 2\pi L\Delta f/c,$$

where Δf is the width (FWHM) of the resonance curve.

The spectrometer uses the technique for extracting the instrumental function by the replacement of the absorbing sample in the resonator with a nonabsorbing one (e.g., pure nitrogen). In this case, the energy losses in the sample are zero, and the resonator losses (the instrumental function or base line) may be defined as

$$P_{\text{res}} = 2\pi L\Delta f_0/c,$$

where Δf_0 is the width of the resonance curve with zero losses in the sample.

Hence, the losses in the sample are

$$P_{\text{samp}} = 2\pi L(\Delta f - \Delta f_0)/c.$$

The half-width of the resonance curve is computed by the least-squares fitting of the resonator response to the known theoretical profile. The data of each scan are a mixture of the useful signal, the noise, and the spurious signal. The useful signal is described by the Lorentzian function. The spurious signal is a result of radiation interference in the channel and, in view of the narrow scanning range, may be approximated by a linear function. The noise signal that corresponds to the noise level of the detector and the residual bias voltage of the amplifier may be represented by an additive frequency-independent term. The resultant function describing the total signal is

$$F(f) = \frac{A + B(f - f_0)}{(\Delta f/2)^2 + (f - f_0)^2} + C,$$

where f_0 is the resonant frequency and A, B, and C are the parameters of the spurious signal (A and B include the useful-signal amplitude).

The data processing consists in finding the coefficients of this function for each scan.

The scans are processed separately, and the results of processing are averaged over all of the scans to increase the SNR. Two averaging methods may be used: the averaging of the widths obtained for each scan or the matching of the centers of the recorded profiles followed by their averaging and determination of the half-width of the averaged line. The advantage of the first method is that it is rapid. The second method provides a possibility of monitoring the purity of the experiment. If the setup is exposed to an external action (e.g., shaking, movement of the air, etc.), the shape of the recorded line is distorted. The presence of distorted profiles among those being averaged is detected by a bending of the residual (a difference between the experimental data and the theoretical function); a flat noise-locking residual indicates that the data are correct.

The result of processing 500 scans is shown in Fig. 2 to illustrate the use of the second averaging method. The residual of the fit of the experimental record by the Lorentz profile, which is presented at the bottom of Fig. 2, demonstrates the adequacy of the model in use and the high (~400) SNR. The width of the resonance curve in this record determined from the processing is Δf = 164728 (20) Hz. The measurement accuracy is only 20 Hz. This means that the minimum change in the absorption that can be detected by the spectrometer is $\sim 4 \times 10^{-9}$ cm^{-1}.

The frequency dependence of the absorption in the sample is obtained by processing the results of scanning at the resonator's eigenmodes in the range under study. In this case, the absorption factor γ is found from the formula

$$P_{\text{samp}} = 1 - e^{-\gamma L} = \gamma L \quad (\text{at } \gamma L << 1). \tag{2}$$

A record of the atmospheric spectrum in the range of 110–205 GHz is shown in Fig. 3 to illustrate the broadband study of the absorption in gases. Each point

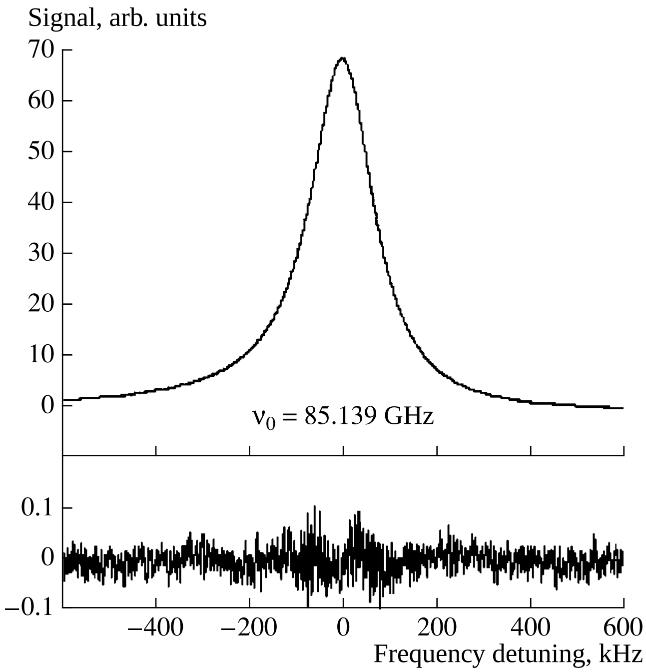

Fig. 2. The resonance curve with a center frequency of 85.139 GHz (at the top) and the difference between the experimental record and the result of its processing with a Lorentz profile (at the bottom).

corresponds to the frequency of the resonator's eigenmode. A solid line presents a fit of the theoretical Van Vleck–Weisskopf profile to the observed atmospheric lines of H_2O at a frequency of 183310 MHz and O_2 at a frequency of 118750 MHz. Constant, linear, and quadratic (in frequency) terms were added to the theoretical line shape to take into account nonresonant atmospheric absorption. At the bottom, the figure presents the difference between the experimental record of the line at atmospheric pressure and the result of its processing, which demonstrates the adequacy of the selected theoretical shape.

An advantage of this equipment is its ability to measure the absolute value of the absorption in a gas. A polyethylene cap that seals the resonator cavity against ambient conditions is used both to measure the instrument function and to study the absorption at different concentrations of the absorbing gas. This has made it possible, in particular, to record the line of water at different humidity values (Fig. 4) and determine the parameters of the line (the center frequency and the parameters of broadening and shift by dry air, nitrogen, and oxygen) with a high accuracy [6].

Similar measurements for the oxygen line at ~118 GHz allowed the parameter of the spectral-line interference with lines from the absorption band at 60 GHz to be measured in a laboratory experiment. It is important to note that this parameter, required for processing the results of remote atmosphere sensing, has never been measured in a direct experiment but was predicted from the experimental results for other lines. At the same time, neglect of this parameter introduces an error into the reconstruction of the atmospheric parameters from the results of the remote sensing.

4. DISCUSSION

For now, the minimum time it takes for an absorption spectrum of a sample to be measured over the entire operating frequency range of the BWO may be estimated as follows. The minimum time between the frequency changes due to the fast synthesizer is 58 μs;

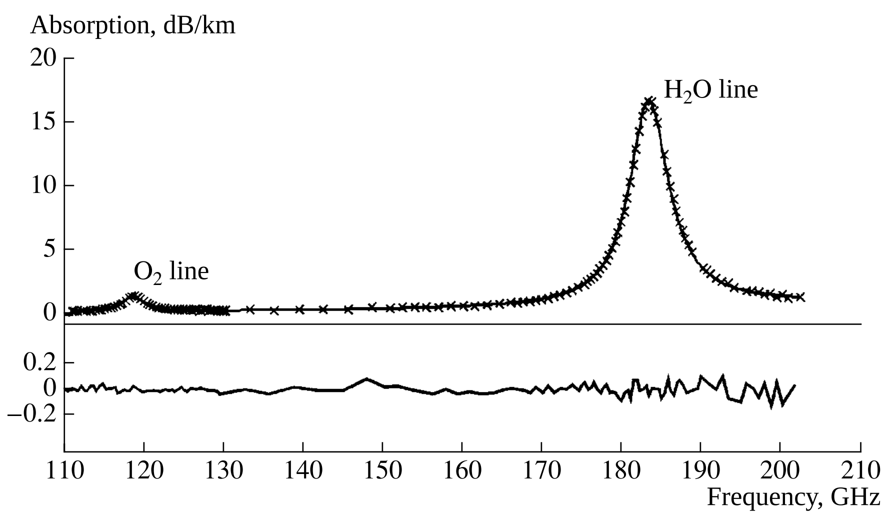

Fig. 3. The top: the spectrum of the open atmosphere for 110–205 GHz with absorption lines of oxygen (118 GHz) and water (183 GHz). The temperature is 24.5°C, the pressure is 756 Torr, and the humidity is 4.75 g/m^3. The bottom: the difference between the experimental record and the theoretical Van Vleck–Weisskopf model.

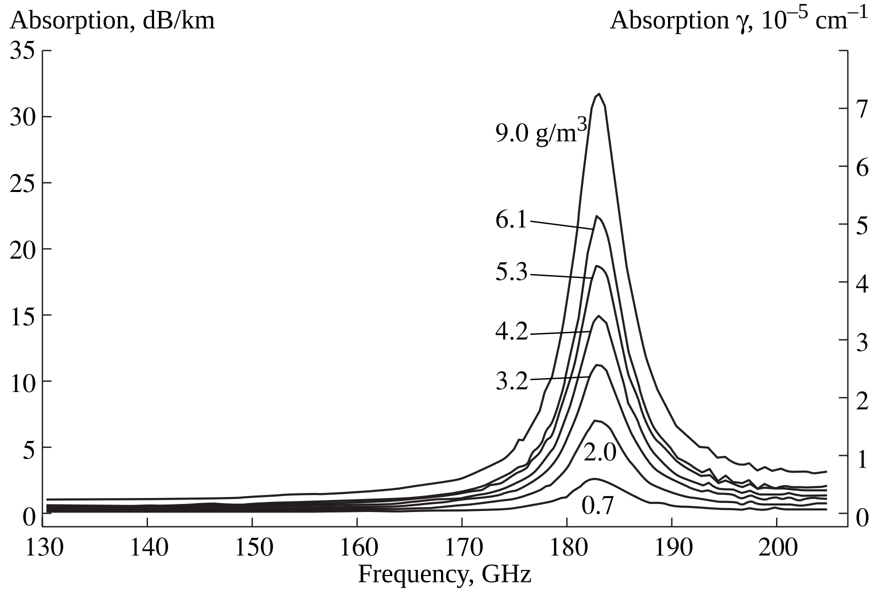

Fig. 4. The record of the water line of 183 GHz at atmospheric pressure, room temperature, and different values of humidity.

a single record of the forward and backward scans with 100 points each takes ~11.6 ms. A change between the resonator modes in view of the time required for the tuning to a frequency of the MW synthesizer and the BWO takes ≈40 ms; i.e., ~4 s is needed for the frequency change over the entire frequency range comprising ~100 modes. The total data-recording time is 1.16 s. Twenty thousand readings recorded during the forward and backward scanning at all of the modes are transmitted to the PC memory in ≈0.96 s. The processing time depends on the SNR in the record of the response and, on the average, is <1 s. As a result, ≈6 s passes between the start of the experiment and the display of the resultant spectrum for the entire frequency range of the BWO on the PC screen. This means that the information on the absorption in the sample over a ~100-GHz range can be obtained virtually in the real-time mode. A change in the absorption in the sample, e.g., while monitoring the humidity during an industrial production process, may be detected within a significantly shorter time. The measurements can be carried out at the maximum absorption of the water line without changing to another mode. In this case, the spared time may be used to accumulate the signal, thus increasing the sensitivity of the instrument and the measurement accuracy.

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