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MODERNIZATION OF THE KHARKIV MICROWAVE SPECTROMETER: CURRENT STATE

Subject and Purpose. *Results are presented of the recent considerable upgrade implemented at the Kharkiv microwave spectrometer. The upgrade has been aimed at extending the operating frequency range and increasing the utmost accessible spectral resolution of the spectrometer.*

Methods and Methodology. *In order to extend the frequency range we have designed and constructed new BWO-based oscillator units, also providing for possibility of frequency tripler application. Construction of a new absorbing cell of enlarged diameter allowed us to considerably improve the spectral resolution for Lamb-dip measurements.*

Results. *Owing to the upgrade, the spectrometer has become able to cover the frequency range from 34 to 420 GHz, with a gap from 183 to 200 GHz. The spectral resolution in the Lamb-dip observation mode has been increased by a factor of two. In addition, the functionality of the spectrometer has been significantly improved via modernization of several of its subsystems.*

Conclusions. *The new upgrades of the spectrometer systems have permitted extending the operational frequency range and increasing the utmost accessible resolution by means of reducing the time-of-flight line broadening in the Lamb-dip measurements. In addition, application of square-wave frequency modulation with accurately known modulation parameters, in combination with careful modeling of the distortions caused by reflections in the absorbing cell, has allowed us to significantly improve the accuracy of line frequency measurements.*

Keywords: *microwave spectrometer, millimeter wave spectrum; measurement accuracy, spectral lines.*

Introduction

Rotational spectroscopy is a powerful method which is in demand in a number of scientific disciplines, including molecular physics, physical chemistry, radio astronomy, etc. The relationship between the molecular structure and the rotational transition frequencies, and such effects as centrifugal distortion, nuclear quadrupole coupling, additional splittings

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caused by large amplitude motions, etc. provide useful information on the structure and internal dynamics of the molecules under study. The continuous development of experimental techniques, as well as upgrades of instrumentation in the field of microwave spectroscopy underlie the progress achieved in the study of various scientific problems, opening new directions of research.

In this paper we present results of an upgrade of the Kharkiv microwave spectrometer [1, 2], which has been successfully employed, for quite a long time, for high-accuracy investigations of molecular rotational spectra (within the research programs of the Institute of Radio Astronomy, NAS of Ukraine — see, for example [3—7]). The most recent description of the spectrometer was presented almost a decade ago [2], while a number of new results were briefly reported in the conference paper [8]. In the present paper, we would like to summarize the results of our permanent efforts aimed at improving the spectrometer and describe the current state of our main scientific instrument.

As is known, performance of a spectrometer may be described by the parameters as follows:

- Operating frequency range.
- Spectral resolution, i.e. the minimal difference in frequency between two closely lying spectral lines which can be measured individually by the instrument.
- Accuracy of measurements, i.e. estimated error of frequency determination for central frequencies of molecular spectral lines.
- Sensitivity, which is determined as the absorption coefficient of the weakest spectral line detectable by the instrument.
- Productivity, i.e. the time interval needed to record a molecular spectrum within a particular frequency range.

During the last decade, the efforts for modernizing the Kharkiv microwave spectrometer [1, 2] were focused on improving the three first parameters from the list above. The tasks included extension of the operating frequency range from 49—250 GHz to 34—420 GHz and design of a new absorption cell of an enlarged diameter, such as to allow increasing the spectral resolution for sub-Doppler measurements via Lamb-dip observations. In addition, development of a square-wave frequency modulator system, combined with the new approach as to taking into account modulation distortions and the distortions caused by standing waves in the absorption cell, allowed improving the measurement accuracy quite significantly. The details of our new approach to accuracy improvement were recently reported in paper [9]. Here, we will discuss, in some more detail, the technical solutions which have allowed us to implement the square-wave frequency modulation with accurately known parameters. In addition, the extension of the operating frequency range involved modernization of the phase lock loop (PLL) system and of the spectrometer receiving system. Details of modernization of these subsystems will be also discussed in this paper.

The rest of the paper is organized as follows. Section 1 represents a brief description of the block diagram of the Kharkiv microwave spectrometer, aimed at suggesting a general framework for discussions of the upgrade that has been undertaken. In Section 2 we discuss the ways for extending the operating frequency range and the accompanying modernization of some of the spectrometer systems. In Section 3 we consider the spectral resolution improvement owing to employment of the new absorption cell. The Conclusions summarize the results obtained in this work.

1. Block-Diagram of the Improved Spectrometer

The automated synthesizer-based, millimeter/submillimeter wave spectrometer of the Institute of Radio Astronomy, NAS of Ukraine belongs to the class of conventional absorption spectrometers. It involves a set of backward wave oscillators (BWO) covering a frequency range from 34 to 183 GHz, with allowance for further extension to 200—420 GHz with the help of solid state passive frequency multipliers. A simplified block-diagram of the spectrometer which reflects the recent upgrades of different subsystems is shown in Fig. 1. The 34 to 420 GHz radiation from the microwave frequency synthesizer (see Fig. 1) passes through the absorbing cell which contains the gas to be investigated, and is fed into the Schottky detector and further on to the receiving system. The microwave frequency synthesizer is built on the basis of phase-locked BWOs. The reference signal of 385—430 MHz for the microwave frequency synthesizer is provided as an up-converted output signal from the direct digital synthesizer (DDS) AD9851. The long-

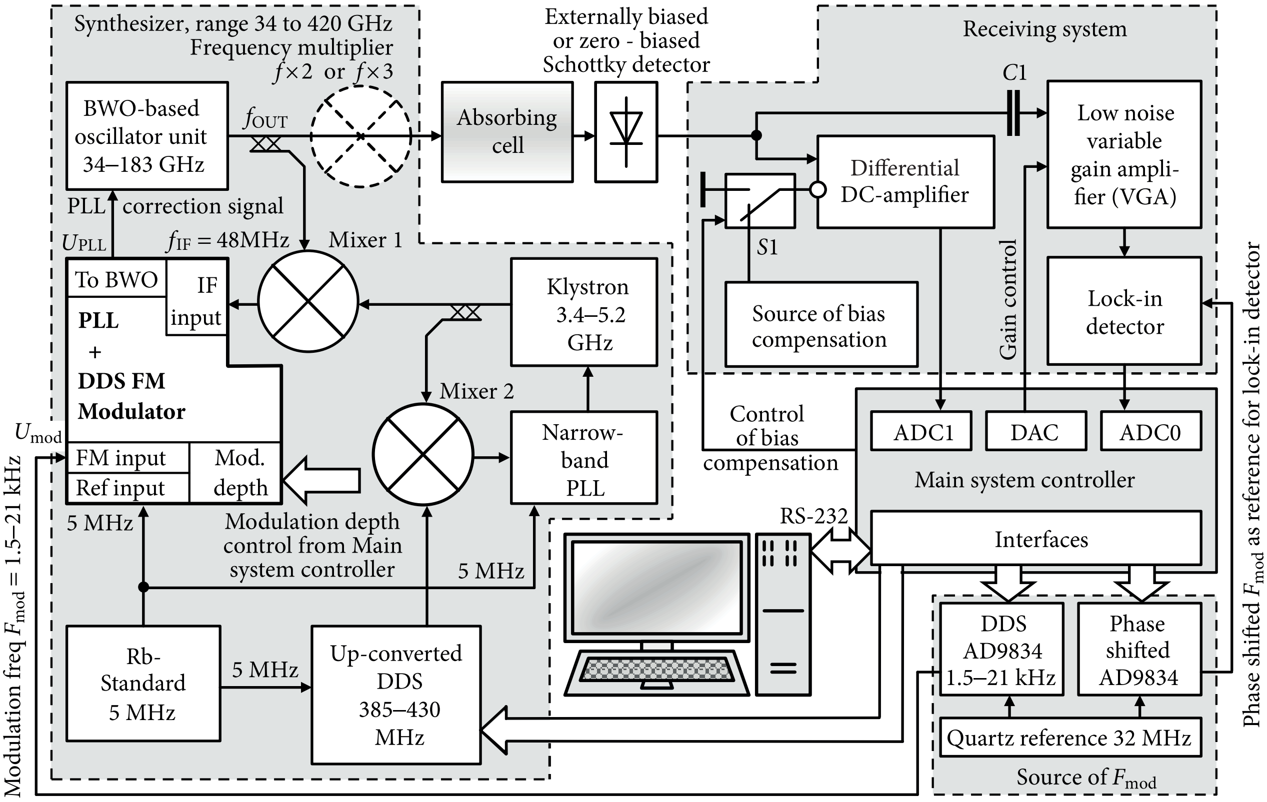

Fig. 1. Block-diagram of the Kharkiv spectrometer (current state). The structure of the PLL+DDS FM Modulator unit is presented in detail in Fig. 2

term frequency stability is determined by the stability of the rubidium frequency standard. The spurious components of the DDS signal (this problem was earlier discussed in detail in [1, 10, 11]) are filtered at the first stage of frequency multiplication by means of a narrow-band PLL (pass-band about 1 kHz). That provides phase stabilization for the signal from a klystron operating within the range of 3.4 to 5.2 GHz [1, 2]. At the next stage of frequency multiplication the millimeter-wave BWO is phase locked (via the PLL+DDS FM modulator, see Fig. 1) to harmonics of the klystron. To cover the frequency range of 34—183 GHz we apply a number of BWO-based oscillator units. Further extension of the frequency range, up to 420 GHz, is achieved by means of passive frequency multipliers. In order to improve sensitivity, the output signal of the radiation source (phase-stabilized BWOs) is frequency modulated and lock-in detection is applied in the receiving system. The spectrometer hardware is connected to the control computer by means of the main system controller (see Fig. 1), via a standard serial interface RS-232.

The spectrometer operates as follows. The frequency modulated (FM) radiation from the stabilized BWO, of a carrier frequency f_{OUT} (otherwise, $2f_{OUT}$ in the case of application of a frequency doubler, or else $3f_{OUT}$ in the case of a frequency tripler, see Fig. 1) passes though the absorbing cell which contains the sample under study, to be received by a room temperature Schottky detector. The output signal from the detector, with a modulation frequency F_{mod}, is amplified in a low-noise, variable gain amplifier (VGA, see Fig. 1) and measured, upon lock-in detection, in the analog-to-digital converter ADC0.

The probing signal of the spectrometer is provided by a PLL-stabilized BWO and our modulation scheme is based on application of a frequency modulated reference signal for that PLL. The FM signal is produced in the DDS providing frequency-shift keying (square-wave FM) between two frequencies that are known to a very high accuracy (see the PLL+DDS FM modulator in Fig. 1). Such an approach has allowed us controlling parameters of the frequency-modulated signal to a high accuracy, which is of impor-

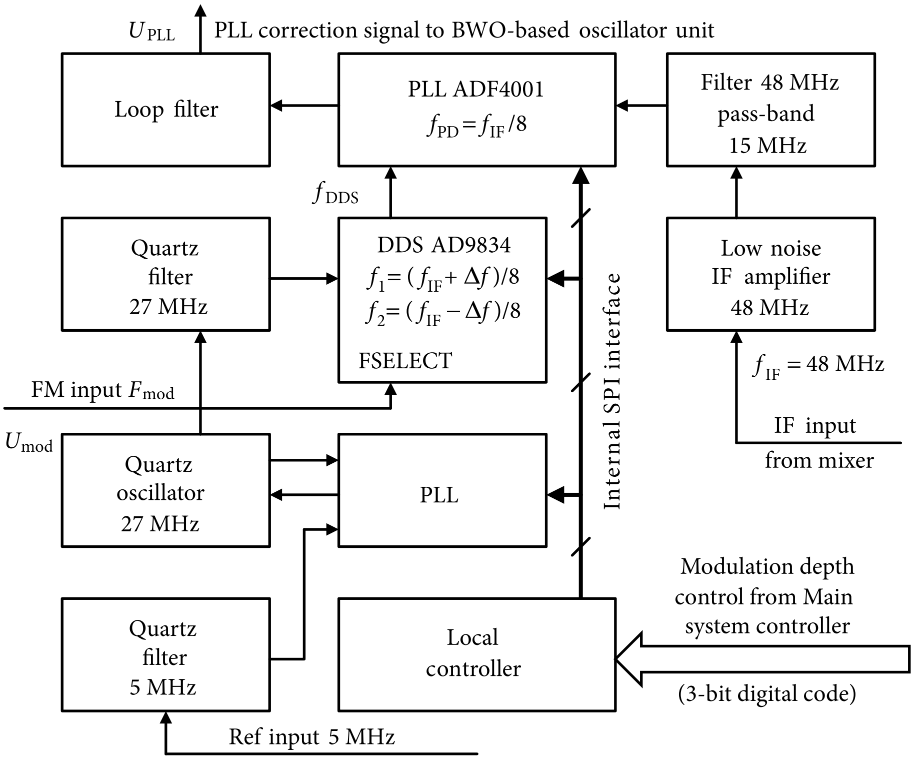

Fig. 2. Block-diagram of the PLL + DDS FM modulator (see Fig. 1)

tance for obtaining the highest possible accuracy of line frequency measurements. The approach has already been presented in paper [2], so we are discussing here some technical details and further developments. We note in passing that the theoretical approach which allows taking into account the modulation- and standing wave-produced distortions in the case of square wave modulation was discussed in detail in paper [9].

A simplified block-diagram of the modulator is shown in Fig. 2. The FM signal is generated by means of the digital synthesizer (DDS) AD9834 [12]. As is known, the spectral purity of the signal at the DDS output is strongly dependent on quality of the relevant reference signal. For this reason we applied multi-stage reference filtering. First, the signal from a rubidium standard (see Fig. 2) is filtered by a narrow band quartz filter (central frequency equal to 5 MHz, pass-band about 1 kHz). This filtered signal is used as a reference for the PLL which stabilizes the 27 MHz quartz oscillator whose signal, in its turn, is subjected to filtering by an additional narrow band quartz filter. The approach allowed us to improve the spectral purity of the DDS reference signal and, accordingly, such of its output signal.

In order to obtain a frequency modulated reference signal, the DDS is programmed by a local controller, via an internal Serial Peripheral Interface (SPI, see Fig. 2), so as to provide two frequency profiles, f_1 and f_2. The logic-level modulation signal U_{mod} at a frequency F_{mod} serves to periodically switch the output frequency of the DDS (FSELECT input of AD9834, see Fig. 2) between two values, $f_1 = (f_{IF} + \Delta f)/8$ and $f_2 = (f_{IF} - \Delta f)/8$. Here f_{IF} is the intermediate frequency (IF) of the PLL system stabilizing the BWO frequency; Δf stands for modulation depth (see Fig. 3, the meaning of the factors 1/8 in f_1 and f_2 will be discussed below).

The magnitude of the modulation depth Δf may be selected from among the values 1, 2, 4, 8, 16, 32, 64, or 128 kHz. The switching frequency $F_{mod} = 1/T_{mod}$ is formed by an additional DDS (Source of F_{mod}, see Fig. 1). In order to obtain both the modulating signal and the reference for the lock-in detector, we applied two DDSs operating with the same frequency F_{mod} but with controlled phase shift (the AD9834 DDS for 1.5—21.0 kHz and the Phase shifted AD9834, see Fig. 1). Depending on experimental conditions the modulation frequency F_{mod} may be selected within the range 1.5—21.0 kHz.

The square-wave frequency-modulated signal with accurately known parameters, obtained as described, is applied as a reference for the ADF4001 PLL (see Fig. 2) which stabilizes the frequency of the signal from the BWO, providing its FM output. The necessary parameters of the PLL are provided by a corresponding loop filter (see Fig. 2). So, molecular spectra are recorded with the aid of a square-wave FM radiation source.

In order to exploit advantages of the new modulator with accurately known modulation parameters one should develop a corresponding approach to properly process the records. Accuracy of line frequency measurements is one of the most important parameters for spectrometers. The central frequencies of spectral lines are usually determined by means of a least squares fit applied to a record, with a properly selected function for the line profile. However, it is well known [13] that the line shapes recorded are affected by the distortions owing to modulation and standing wave effects.

As a result, the accuracy of frequency measurements may happen to be considerably decreased. Since our previous approaches to data processing were intended for analyzing the line shape records as obtained with a sine-wave FM, we have faced the need for developing a new approach that would allow taking into account peculiarities of the square-wave FM. The new approach allows one taking into account both the modulation-induced distortions and the distortions due to standing waves. This provides an opportunity for improving the accuracy of line frequency measurements by approximately an order of magnitude. The details of this approach, as well as the test results concerning the improvements achieved can be found in paper [9].

2. Extension of the operating frequency range

As mentioned in the Introduction, extension of the operating frequency range of the spectrometer has been one of the goals of this modernization. Previously, the spectrometer covered a frequency range from 49 to 149 GHz [2]. In total we applied three different oscillator units (covering 49—84 GHz, 73—119 GHz, and 110—149 GHz) based on three BWOs. When we needed to measure, for radio astronomical purposes, the spectrum of glycine [14, 15] we performed the first in order of appearance extension of the frequency range with the help of a passive frequency multiplier. Application of a home-made frequency doubler and its corresponding Schottky detector allowed us to additionally cover the frequency range from 200 to 250 GHz. So, by the time we have started the present stage of spectrometer modernization the lowest operating frequency of the spectrometer was 49 GHz, while the highest one equaled 250 GHz with a rather wide frequency gap between 149 and 200 GHz.

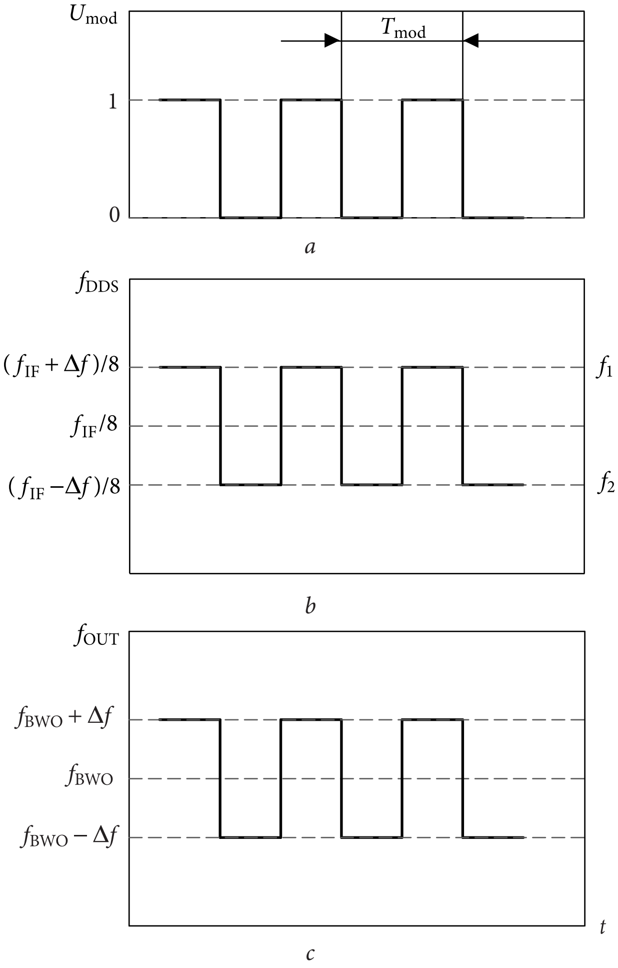

Fig. 3. Time diagram of the FM signal. The modulation depth has been denoted as Δf, and modulation frequency is $F_{\text{mod}} = 1/T_{\text{mod}}$, where T_{mod} stands for the period of modulation (*a*). The DDS output frequency has been chosen to be $f_{\text{DDS}} = f_{\text{IF}}/8$. The reason for dividing by 8 of the f_{IF} PLL frequency arises from performance limitations of the AD9834 which is not capable of generating 48 MHz. Since the PLL reference signal f_{DDS} is frequency modulated (*b*), the f_{OUT} also appears to be frequency modulated (*c*)

Ideally, the operating frequency range of a spectrometer should be coordinated with the problems to be solved. While extension of the range to a scale as wide as possible (aimed at covering all the relevant frequencies) might be desirable in theory, in practice the operating frequencies are limited in magnitude by the performance characteristics of the oscillation sources available. One of the important applications of the microwave spectroscopy in general and of our spectrometer in particular is providing reference spectroscopic information for radio astronomy studies of the interstellar medium. Nowadays the Atacama Large Millimeter/submillimeter Array (ALMA) is one of the cutting-edge instruments applied for radio astronomy of the Universe, including studies of the interstellar molecular clouds. These studies need to be supported with corresponding reference spectroscopic data from a laboratory.

At present, the ALMA covers the frequency range from 35 to 950 GHz, with certain gaps in between [16], while the majority of spectral surveys of molecular clouds still do not go above 400 GHz. In the framework of our upgrade of the spectrometer one of the goals was to achieve a wider band of matching with the operating range of the ALMA telescope. As is known, the low frequency part of the spectrum is the domain where the spectra of rather heavy polyatomic molecules may be observed. Actually, this is the reason why a 35 to 50 GHz range receiver has been manufactured and installed in ALMA's antennas. Moreover, the first case of reception of an astronomical signal has recently been reported for this band [17]. That is why our efforts on modernizing the spectrometer have been aimed at extending the operating frequency range both down and up in frequency.

The extension of the frequency range was achieved by means of incorporating in the spectrometer of additional BWO-based radiation sources, as well as by application of an additional frequency multiplier. The new radiation sources are represented by two new oscillator units which we have designed and constructed, based on the backward wave oscillators OB-69 and OB-87. The former one (covering the range of 34—56 GHz) allows extending the frequency range downward to 34 GHz. The latter covers the frequency range from 115 to 183 GHz. Application of this unit has allowed us to noticeably reduce the previously existing frequency gap, namely from 51 GHz (i.e., between 149 and 200 GHz) down to 17 GHz (183—200 GHz).

The main feature of the BWO is that its frequency response is dependent on the anode voltage, and hence the BWO is a voltage controlled oscillator with nearly ideal properties. A BWO-generated frequency can be tuned within the full frequency range by changing the anode voltage, without moving any mechanical parts. For this reason the full-range control over the BWO frequency can be provided by a remotely controlled high-voltage power supply. A block-diagram of the BWO units is shown in Fig. 4. The structure of the two new units is rather similar. The major differences between the two units are the type of the BWO and the range of its high-voltage power supply (cf. 400 to 1 400 V for the OB-69 and 800 to 2 800 V for the OB-87). Each of the oscillator units involves a BWO, a high-voltage regulator, and sources of the filament and grid feed voltages. It should be noted that both the anode of the BWO and its waveguide output are grounded, while the rest of BWO's electrodes and power supplies (like the filament current and the grid voltage regulators) are connected to the negative high voltage bus as is shown in Fig. 4. The voltage level at the output of the high-voltage regulator is controlled by its reference voltage U_{ref} (variable between 0 and 2.5 V) provided by the digital-to-analog converter (DAC) which is an internal part of the micro-controller ADuC841 (see Fig. 4). At the same time this micro-controller provides for remote control over the oscillator unit. Application of the ADuC841 offers an additional advantage as the BWO calibration data have been included to the micro-controller's software. This permits visualizing the current frequency in the 6-symbol, 7-segment LED display (see Fig. 4, BWO frequency display). At the same time manual control of the output frequency is also provided.

The highest spectral resolution and measurement accuracy of the spectrometer can be achieved only through application of frequency synthesis. For this purpose the BWO of oscillator unit should be synchronized by means of an external PLL using a frequency synthesizer as a reference. The main problem for such synchronization is that the BWO operates at rather high anode voltages (up to 2 800 V), such that the PLL correction signal U_{PLL} characterized by a bandwidth of few megahertz (with the DC included) should be added to the voltage from the anode power supply. As is known, the pass-band of a power supply usually is quite limited (making just a few kilohertz).

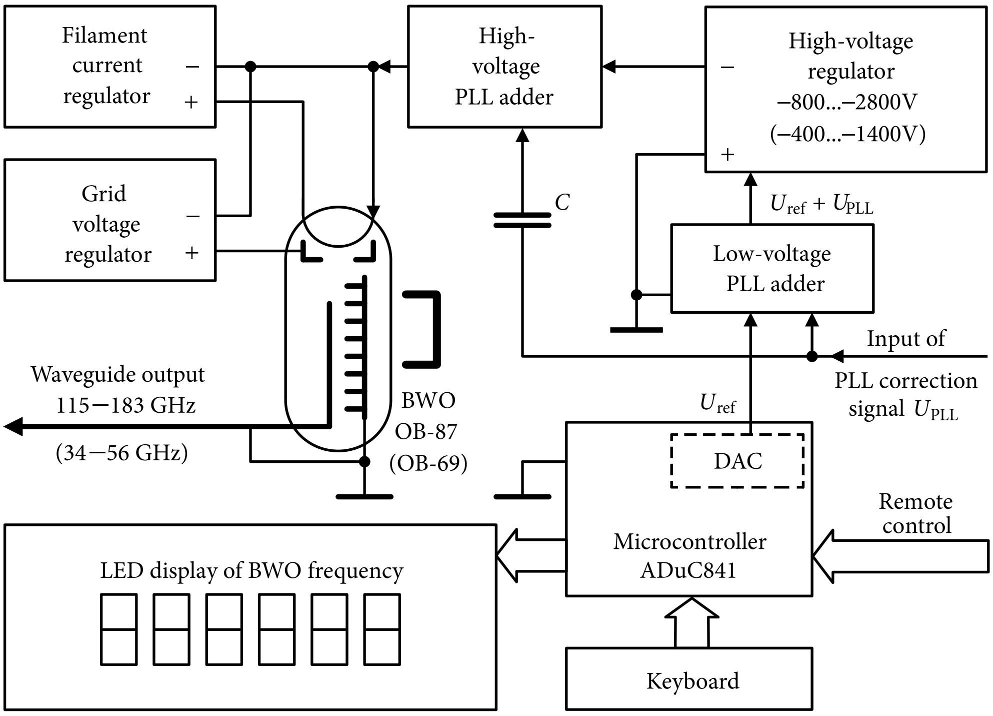

Fig. 4. Block-diagram of oscillator units: versions for frequency ranges 115 to183 GHz and 34 to 56 GHz. The two units are of rather similar structure, differing only in the type of their BWOs and some voltage parameters of their power supplies. The parenthesized designations relate to the BWO OB-69, while those unbracketed to OB-87

In order to provide correction within the pass-band of a few megahertz we have equipped the oscillator unit with a high-voltage PLL adder; a capacitor *C*, and a low-voltage PLL adder (see Fig. 4). Thus, the correction signal U_{PLL} ranging in frequency from DC to approximately 30 Hz is added to the reference voltage U_{ref} obtained from DAC. The correction signal above 30 Hz passes directly through the capacitor *C* (see Fig. 4) to be fed into the high-voltage adder. This is how the wide-band correction signal is provided for the BWO's anode power supply.

The new oscillator units were incorporated into the existing system of frequency synthesis, so the output frequency of these units is synchronized by the rubidium standard. An example of operation of the spectrometer with a 34 to 56 GHz low frequency oscillator unit is shown in Fig. 5, where a fragment of molecular spectrum of the dimethyl ether molecule is presented. The fragment for a different (higher-frequency) range, namely the one obtained with the 115 to183 GHz oscillator unit is shown in Fig. 6.

The significant expansion of the operating frequency range of the spectrometer has lead to considerable variations in the level of the f_{IF} signal from the PLL which is used to stabilize the BWO output frequency. Usually, a rather high IF-signal was observed in the case of the low-frequency BWO (with a risk of locking to the second harmonic of the IF-signal), whereas a too small IF-signal was obtained in the case of the high-frequency one. For this reason the PLL system was significantly reconstructed in comparison with the previous variant [1, 2].

First of all, we have increased the maximum gain of the IF-amplifier, at the same time providing an opportunity for adjusting the gain. The gain variation is about 30 dB. This allowed us to optimize the gain required for the best operation of this PLL over the whole operating range. By increasing the maximum IF-amplifier gain we identified some kind of interference between the PLLs of the klystron and the BWO. That is why we decided to change the value of the f_{IF}. The previous values of the f_{IF} were either 25 or 50 MHz [1, 2], hence the interference observed could be attributed to infiltration of harmonics of the 5 MHz klystron PLL's intermediate frequency. Now the intermediate frequency f_{IF} of the BWO's PLL is chosen to be 48 MHz, hence a value not representing a multiple of 5 MHz. Such a solution has allowed us

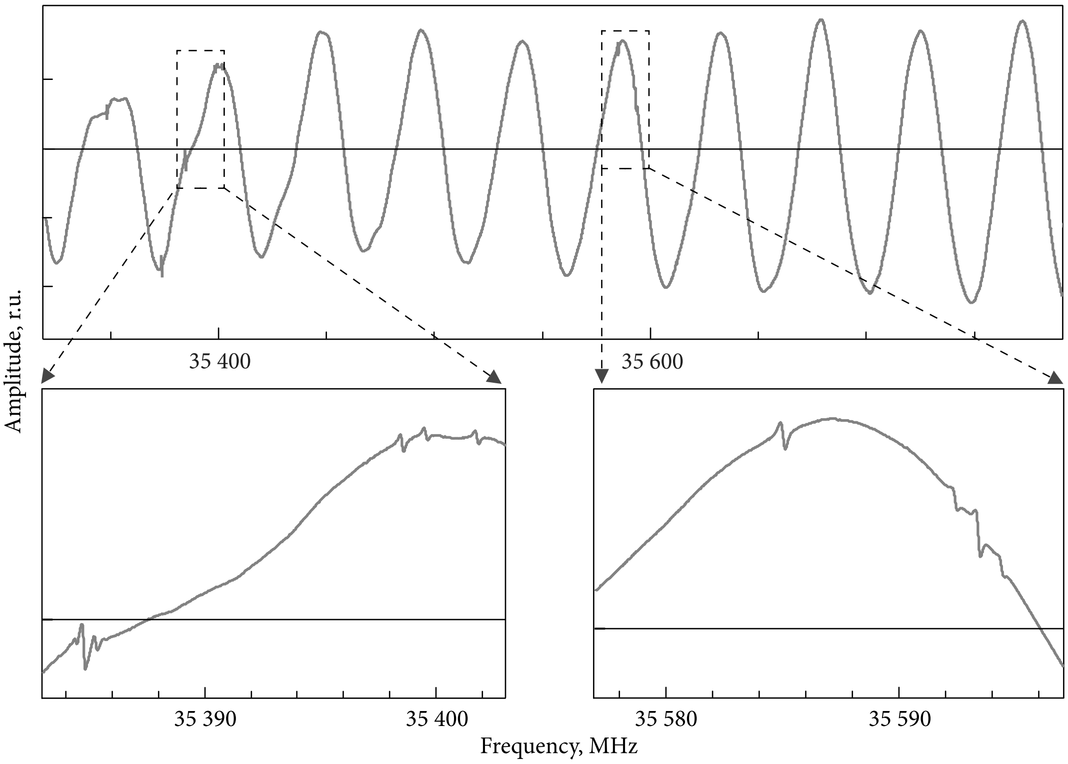

Fig. 5. Fragments of molecular spectrum of dimethyl ether corresponding to the lower frequency end of the spectrometer operating range. The spectra in the bottom line are zoomed views of selected areas on the top

to avoid occurrence of interference between the PLLs of the klystron and the BWO, thus improving reliability of the frequency synthesis. Because of limitations of the AD9834 synthesizer which is not capable of producing a 48 MHz output signal, the operating frequency f_{PD} of the phase detector in the ADF4001 (see Fig. 2) was chosen to be 8 times lower than the PLL intermediate frequency f_{IF}, i.e. $f_{\mathrm{PD}} = f_{\mathrm{IF}} / 8$ (the PLL synthesizer ADF4001 was programmed to perform this frequency division, and the necessary parameters for the PLL are provided by the loop filter as shown in Fig. 2). In order to improve the signal-to-noise ratio of the PLL that stabilizes the BWOs' output frequency, the bandwidth of the IF signal has been limited by application of a band-pass filter (see Fig. 2). To improve the lock-in stability we decided to reduce the filter's pass-band to approximately 15 MHz (compare with the former 100 MHz [1, 2]). It appeared that a pass-band like that was still sufficient for an easy search of the lock-in mode, at the same time, this band-pass value has proven to be able providing for a much better lock-in stability.

Further extension of the operating frequency range was achieved with the aid of a new passive frequency multiplier. The previous practice of applying a home-made frequency doubler and its associated Schottky detector allowed us to cover the frequency range between 200 and 250 GHz. In order to increase the upper limit for the spectrometer operating frequency, we applied the frequency tripler WR2.8×3 and zero bias detector WR2.8ZBD from Virginia Diodes, Inc. According to the technical specifications provided by Virginia Diodes Inc. these devices should provide a guaranteed coverage of the frequency range from 260 to 400 GHz. Meanwhile, it has been revealed in our tests that they can operate in a quite acceptable manner between 234 and 420 GHz. Pumping sources for the frequency tripler have been represented by the existing BWO-based oscillator units which cover the frequency ranges of 73 to 119 GHz and 110 to 149 GHz, respectively. Naturally, the BWOs are included in the frequency synthesis system. An example of a spectrum record obtained with the above mentioned frequency tripler and detector is presented in Fig. 8, *a*.

The incorporation of the frequency tripler and its correspondent WR2.8ZBD detector required modernization of the spectrometer receiving system.

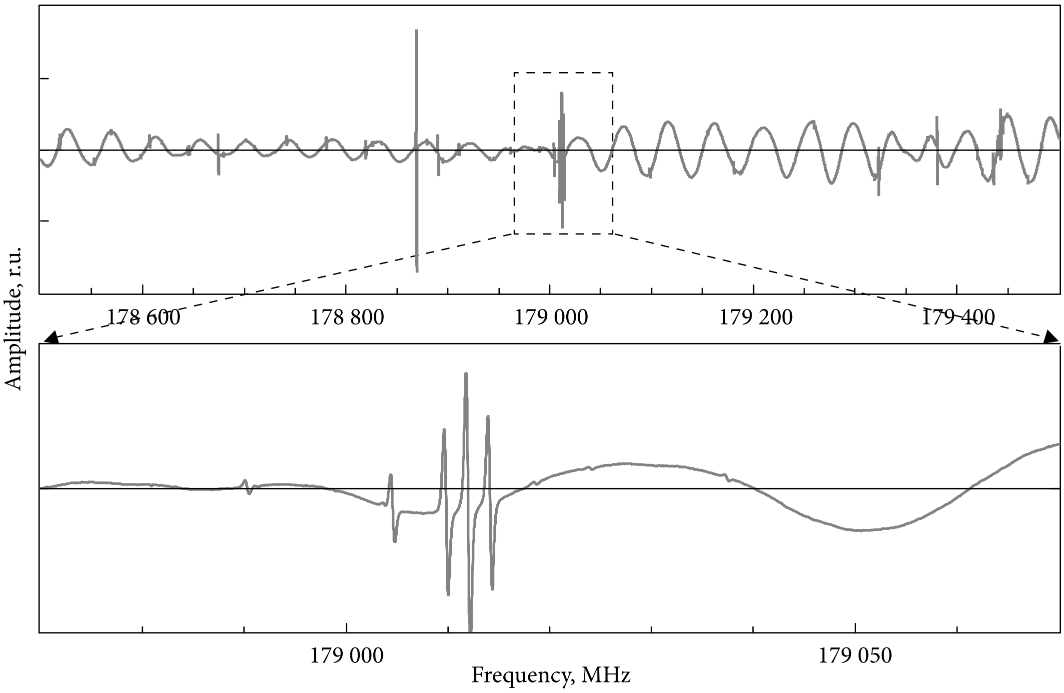

Fig. 6. Fragment of molecular spectrum of dimethyl ether taken around 179 GHz. The spectrum in the bottom line is a zoomed view of a selected area on the top

What we used to apply before were external bias detectors only, whereas the WR2.8ZBD is a zero bias detector. Besides, in the course of several of our latest measuring campaigns we often happened to perform measurements with sub-Doppler resolution, which technique requires readjustment of the time constant of the lock-in detector in the receiving system. Until recently, the time constant of the lock-in detector was fixed at a value which was optimal solely for measurements with a Doppler-limited resolution. So, it seems desirable to develop a new receiving system which would allow easy adaptation to different conditions of the experiment, including quick switching between different kinds of the detectors applied.

In order to meet the requirements put forward in the preceding sub-section, we have developed a new, considerably improved and more efficient receiving system. The system is very adaptive and implements the two-channel technique of spectral recording. One of the channels is a usual alternating current (AC) channel where an AC signal coming from the Schottky detector at the frequency F_{mod} is passed through the capacitor C1 (see Fig. 1) and then amplified in the low noise, variable gain amplifier (VGA, see Fig. 1). The output signal from the channel is measured, upon lock-in detection, with the analog-to-digital converter (ADC0, see Fig. 1). This channel is used for performing high sensitivity recording of the first derivative of the spectrum (see paper [9] for a detailed discussion of the line shape distortions resulting from frequency modulation and lock-in detection). The gain of the VGA depends on the control voltage which is produced by the digital-to-analog converter (DAC, see Fig. 1). The possibility of varying the DAC output voltage from 0 to 2.5 V allows increasing the VGA gain up to +45 dB. Thus, the total gain of the receiving system can be optimized remotely.

It is well known that application of a frequency modulated source with lock-in detection is not suitable for full power measurements of the radiation passing through the absorbing cell. Meanwhile, estimates of this power level are necessary for evaluating absorption coefficients of spectral lines, thus allowing for reducing ambiguity in molecular transition assignments. In order to have an opportunity of measuring the level of the full power passing through an absorbing cell, we have included one more channel to the spectrometer's receiving system. This channel involves a differential direct current amplifier (DC-amplifier) whose output signal is measured by the additional analog-to-digital converter (ADC1, see Fig. 1). The use of a differential amplifier allows applying both externally biased and zero-biased detectors. In the first case the analog switch *S*1 (is also shown in Fig. 1) allows turning on the bias voltage compensation at the input of the differential DC-amplifier. If the zero-biased detector is used, the bias voltage compensation is turned off. An additional useful feature of the new receiving system is the possibility of selecting the time constant of the lock-in detector. Actually, in order to suppress the voltage at the modulation frequency after the lock-in detector we apply third-order active filters with setting times of either 10 ms or 100 ms. This allows us to perform Doppler-limited measurements at a rate of 10 ms per point, while the Lamb-dip measurements are carried out with the rate of 100 ms per point.

To summarize, we note that the operating frequency range of the spectrometer has been extended by more than a factor of two due to incorporation of two new BWO oscillator units and of a passive frequency tripler. The extension of the operating frequency range required a significant modernization of the BWO's PLL system, as well as of the spectrometer's receiving system. As a result of the latest improvements, the spectrometer has become able to cover the frequency range from 34 to 420 GHz, with a 183 to 200 GHz gap in between. Now we are working toward development of an additional oscillator unit to cover the gap.

3. Improvement of spectral resolution

Nowadays, the majority of experiments in the field of microwave molecular spectroscopy are performed with a spectral resolution limited by the Doppler broadening of molecular spectral lines. However, the resolution may be improved if one applies the methods of sub-Doppler spectroscopy with observation of the Lamb-dip [2]. Despite the fact that the technique has some limitations making it poorly suitable for broadband routine studies of molecular spectra, it works very well when the resolution required for measurements of strong molecular lines shall be better than the Doppler-limited level.

Now let us consider the principal limitation for the spectral resolution for sub-Doppler measurements with Lamb-dip observations. Under the conditions of a very low pressure inside the absorbing cell the main factor to limit the resolution accessible in the Lamb-dip mode of measurements is the time-of-flight broadening of the line. As has been shown in [18], the full width of the observed Lamb-dip is dependent on the absorbing cell dimensions as follows:

$$2\Delta\nu_L = 10\left(\frac{2}{D} + \frac{1}{l}\right)\sqrt{\frac{T}{M}}.$$

Here $\Delta\nu_L$ is the Lamb-dip Lorentzian line width in kHz; D and l are, respectively, the diameter and length of the cell (cm); T is absolute temperature (K), and M the molar mass of the molecule (g/mol). Taking into account that the diameter D is much smaller than the length l, the time-of-flight-induced line broadening in Lamb-dip measurements can be reduced through increasing the cell diameter. The former cell, employed in our regular experiments, was made of a glass tube 56 mm in diameter and nearly 3 m in length. Aiming to improve the spectral resolution accessible in the Lamb-dip mode, we have designed and constructed a new absorbing cell, of a 129 mm diameter and a 3 m length. The cell has been made from a stainless steel tube and provided with 6 mm thick vacuum windows made of Teflon©. The additional advantage of the new cell is its lower level of self-leakage, which allows keeping the required level of pressure for a considerable period of time (like several hours) without resorting to continuous pumping. This regime of steady state measurements proves to be especially useful when just a small amount of the sample material is available (like in the case of samples with an enriched isotopologue content). The reduction of the self-leakage rate has been achieved through removal from the cell structure of a number of rubber gaskets (which were part of the cell structure in the preceding version [1, 2]). The new cell is coupled to the radiation source and the detector via home-made dielectric horns. Initially, we tried to use metallic horns, however they showed an unacceptable level of reflections, with the corresponding magnitudes of the standing wave ratio (SWR) reaching a few units. That is why, in an attempt of minimizing the reflections, the horns were produced, at the next stage of modernization, from a fiber-glass plastic with an admixture of a carbon absorbent. Such a solution brought forth considerable improvements as for the situation about the SWR, however at lower frequencies the SWR still remained somewhat above the de-

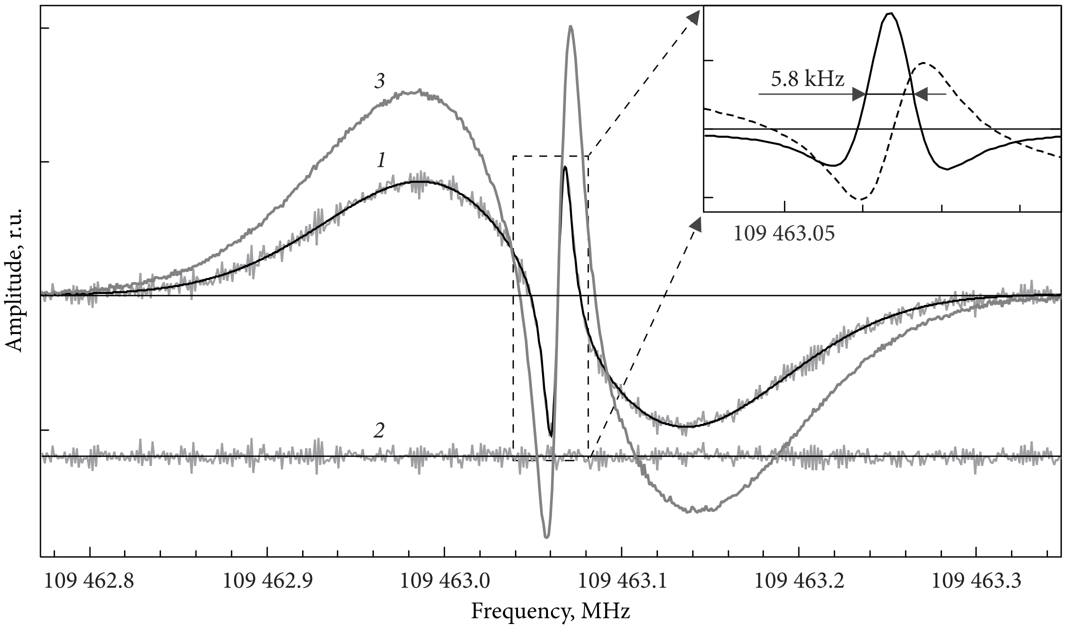

Fig. 7. Lamb-dip record for the $J = 8 - 9$ transition of the carbonyl sulfide (OCS) molecule (measurements performed with an absorbing cell of increased diameter). The Lamb-dip width as obtained from the least-squares approximation of the experimental record is 6.7 kHz. Shown in the insert is the numerically obtained second derivative of the spectrum; full width at half-maximum level is about 5.8 kHz. The value is two times better compared with the earlier result [2], thus suggesting noticeable improvements in the accessible spectral resolution of our spectrometer

sired level. It can be seen from Figs. 5 and 6 that the standing wave ratio is noticeably higher at lower frequencies. In the worst case the SWR at 34 GHz can reach magnitudes about 1.4 to 1.5. However, the value drops down rather quickly at higher frequencies to become lower than 1.05 beyond 38 GHz. To adjust the position of the detector against the field maximum, which might be necessary when the oscillator units are being changed in the process of spectral measurements, the coordinates of the receiving horn may be slightly tuned with four adjusting screws.

The amount of improvement in spectral resolution was tested by recording the Lamb-dip in the $J = 8 - 9$ transition of the carbonyl sulfide (OCS) molecule (the gray colored curve *1* in Fig. 7). The record was approximated by a formula (see the black curve *1* in Fig. 7) using the recently proposed approach [9] which takes into account both the modulation- and standing wave-related distortions. It can be seen that our approach permits a correct reproduction of the observed signal, as the residual difference between the experimental data and the approximation (gray-colored curve *2* in Fig. 7) contains only noise. The measured line frequency, viz. 109463.0641(5) MHz is in good agreement with the 109463.063(5) MHz reported in the literature [19]. The Lamb-dip width obtained from the least-squares approximation of the experimental record is 0.0067(2) MHz (6.7 kHz). Since many of the modern microwave spectrometers combine frequency modulation with second harmonics lock-in detection it is quite popular in the relevant literature to demonstrate the achievable level of spectral resolution by evaluation of the full linewidth values normalized to the second derivative of the Lamb-dip observed. Because of some limitations peculiar to the square-wave FM, our spectrometer is capable of recording the first derivative only. Hence, in order to assess the spectral resolution achieved, with account of normalization to the second derivative of the recorded spectrum, we calculated the second derivative numerically by differentiating the earlier obtained approximating function (the black-colored curve *1* in Fig. 7). The result is shown in Fig. 7 (see the insert in the upper right corner). The full width obtained for the spectrum of the second derivative is about 5.8 kHz at the half-maximum level, which is about two times better than our previous result (10 kHz, see [2]).

The Lamb-dip record (curve *1* in Fig. 7) was obtained with a 2.8 kHz modulation frequency and

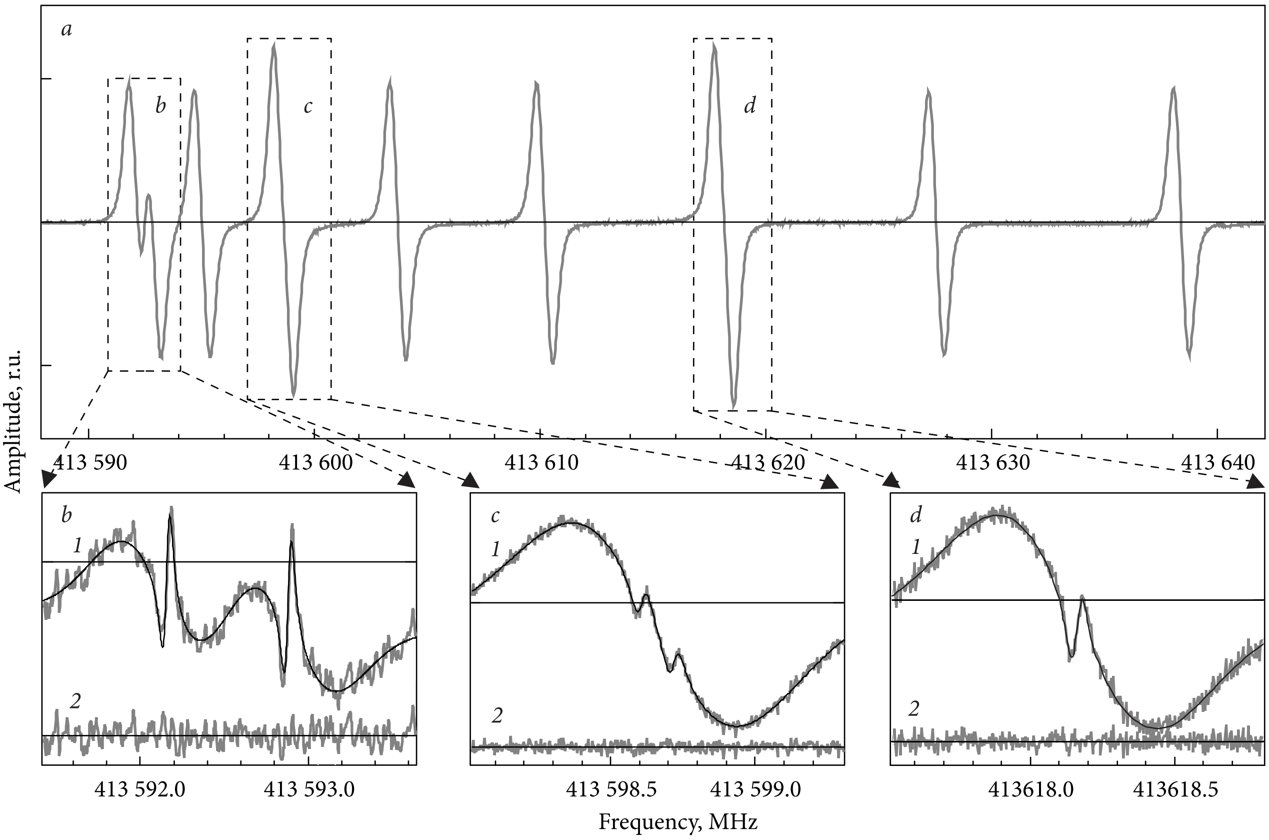

Fig. 8. Parts of the rotational spectrum of CF_3H recorded near 413.6 GHz with the use of the passive frequency tripler from Virginia Diodes, Inc. Several parts of the Doppler-limited-resolution record (*a*) were re-measured with the use of the Lamb-dip technique, see frames (*b*), (*c*), and (*d*). It can be seen that the Lamb-dip technique allowed us to significantly improve the resolution, in particular to resolve the $A_1 - A_2$ splitting in the $K = 3$, $J = 20 \leftarrow 19$ transition of the CF_3H molecule

modulation depth of 2 kHz. The output power of the source was as low as a few micro-Watt, while pressure of the sample was kept close to 1 micro-bar in order to minimize the broadening of the Lamb-dip. The record was obtained as a result of averaging of 159 individual records, which allows us to improve the signal-to-noise ratio (SNR). For comparison, we have also recorded the same line, under the same conditions, but with an increased output power level (the number of records to be subjected to averaging was reduced by one order of magnitude, to 15 individual records). The result is shown as curve *3* in Fig. 7 (scaled to be properly presented in the Figure), and it is clear that, in spite of the significantly reduced time of averaging, the SNR has increased in comparison with curve *1*. Apparently, the record demonstrates a slightly greater Lamb-dip width than in the case of curve *1* which is due to the power-induced broadening effect. Nevertheless, the rather good SNR shown in curve *3* implies that phase noise of the radiation source is of an acceptable level, suggesting an absence of any limitations as for spectral resolution.

As is known, saturation-spectroscopy methods, such as the Lamb-dip technique, are strongly dependent on the level of microwave power in the absorbing cell. Thus, the question remained whether it was possible to perform Lamb-dip observations in the higher-frequency part of our operating frequency range which is determined by properties of the passive frequency multipliers employed. The question is relevant as the output power of passive frequency multipliers is known to make just a few percent of the pumping power (typically, the efficiency of the WR2.8 × 3 tripler is 3% — reported by the Virginia Diodes, Inc. [20]). Fortunately, the rather high output power of the BWOs which are used for pumping the multipliers (specifically, 20 mW or more) and the nearly 3% efficiency of the frequency multiplier, being manifested together, bring forth an acceptable power level of the multiplied signal. This has allowed us performing sub-Doppler measurements in the higher-frequency part of the operating range. Fig. 8 illustrates this capability by presenting records of several transitions of the CF_3H molecule near 413.6 GHz, both for the case of Doppler-limited and the sub-Doppler resolution.

Fig. 8, *a* presents a part of the $J = 20 \leftarrow 19$ *R*-type branch of the CF_3H spectrum recorded with a Doppler-limited resolution. The dashed rectangles (*b*), (*c*), and (*d*) in the main panel mark those lines which were re-measured with a sub-Doppler resolution. Fig. 8, *b* presents a Lamb-dip record of the $K = 0{,}1$, $J = 20 \leftarrow 19$ transitions which were only partially resolved in the Doppler-limited resolution technique. Fig. 8, *c* presents a $A_1 - A_2$ splitting of the $K = 3$, $J = 20 \leftarrow 19$ transition [21] which is not seen at all in the case of Doppler-limited resolution but is well resolved in the sub-Doppler resolution mode. Fig. 8, *d* represents an attempt of observing the $A_1 - A_2$ splitting doublet of the $K = 6$, $J = 20 \leftarrow 19$ transition. The record of Fig. 8, *c* demonstrates an apparent splitting of 0.110 MHz, while the splitting in Fig. 8, *d* is too small to be resolved, even in the Lamb-dip mode. It is supposed that the rest of lines in Fig. 8, *a* should not have any splitting. The solid black lines *1* in Fig. 8, *b* through Fig. 8, *d* represent results of an approximation with the use of the previously described approach [9], while lines *2* show the residual difference between experiment and approximation. As can be seen, these residuals contain only noise, and therefore the quality of approximation can be considered as very satisfactory.

Conclusions

In this paper we have presented results of a significant modernization of the Kharkiv microwave spectrometer that were achieved during the last decade. The upgrades performed have allowed extending the operating frequency range of the spectrometer, improving its measurement accuracy, as well as increasing the utmost accessible spectral resolution through reduction of the time-of-flight line broadening in the Lamb-dip measurements. The development and production of new oscillator units and application of a new passive multiplier have allowed us to extend the operating frequency range up to 34 through 420 GHz, with a small gap between 183 and 200 GHz. Application of a square-wave frequency modulation with parameters known to a high accuracy, complemented by careful modeling of the distortions caused by reflections in the absorbing cell, have considerably (by an order of magnitude) improved the accuracy of line frequency measurements. The new absorbing cell of enlarged diameter has allowed us to reduce the time-of-flight broadening, thus offering a roughly factor of two improvement in the spectral resolution for Lamb-dip measurements.

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МОДЕРНІЗАЦІЯ ХАРКІВСЬКОГО МІКРОХВИЛЬОВОГО СПЕКТРОМЕТРА: СУЧАСНИЙ СТАН

Предмет і мета роботи. Наведено результати нещодавніх суттєвих вдосконалень Харківського мікрохвильового спектрометра. Основною метою цих вдосконалень було розширення робочого діапазону частот і поліпшення максимально доступної спектральної роздільної здатності спектрометра.

Методи і методологія. Заради розширення діапазону робочих частот розроблено та побудовано нові генераторні блоки на основі ламп зворотної хвилі, а також створено можливість застосування потроювача частоти. Впровадження нової поглинальної комірки зі збільшеним діаметром дозволило підвищити максимальну спектральну роздільну здатність при вимірюваннях провалу Лемба.

Результати. Після модернізації спектрометр охоплює діапазон частот від 34 до 420 ГГц із розривом від 183 до 200 ГГц. Спектральну роздільну здатність приладу в режимі спостереження провалу Лемба покращено вдвічі. Додатково, за рахунок модернізації кількох підсистем спектрометра, суттєво покращено його функціональність.

Висновки. Нові вдосконалення систем спектрометра дозволили розширити робочий діапазон частот, а також підвищити максимально доступну роздільну здатність завдяки зменшенню пролітного розширення при вимірюваннях зі спостереженням провалу Лемба. Крім того, застосування частотної модуляції прямокутним імпульсом із параметрами, що відомі з високою точністю, у поєднанні з більш ретельним моделюванням спотворень, котрі можуть спричинюватись відбиттями в поглинальній комірці, дозволило значно підвищити точність вимірювання частот ліній.

Ключові слова: *мікрохвильовий спектрометр, спектр міліметрових хвиль, точність вимірювань, спектральні лінії.*