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2016 J. Phys.: Conf. Ser. 741 012169

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Superconducting integrated terahertz receiver for spectral analysis of gas compounds

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Abstract. A new highly sensitive device for analysis of gas compounds in terahertz frequency range based on the superconducting integrated receiver is being developed. Such receiver for spectral research of Earth atmosphere from balloon-borne instrument was developed earlier in Kotel'nikov Institute of Radio Engineering and Electronics and successfully operated during several flight missions. In this work, the laboratory setup for gas spectroscopy in the range of 450-700 GHz with the noise temperature below 150 K and spectral resolution better than 0.5 MHz is presented. First results of measurements of NH₃ and H₂O absorption spectra are obtained.

1. Introduction

Spectrum analysis of gas compounds in terahertz region (0.1-10 THz) is well known to be very promising method for different applications [1-3]. This is because of huge number of substances having strong absorption lines at THz frequencies. For instance, molecules of H₂O, CO, NO, NH₃, NO₂, CS₂, H₂O₂, which are very important for biology, chemistry and medicine, can be detected by THz spectral analysis. Furthermore, many explosives have «partner» compounds, such as NO, NO₂, NH₃, formaldehyde, acetaldehyde and others, which also have absorption lines in THz range. This makes measurements of absorption lines for detecting gases quite promising technique in the following fields: in medicine for analysis of exhaled air and diagnostics of various diseases indicated by substances-markers, in security systems for detecting dangerous (explosive or poisonous) substances in situ, and others. In addition, high-technology processes in manufacturing fields require controlling the purity of gases and impurities structure and hence can efficiently use spectroscopic method for these aims.

Therefore, development of sensitive devices for gas detecting is actual problem nowadays. We are developing the spectrometer for gas analysis in THz range with high sensitivity, such spectrometer could be used for medical diagnostics of exhaled air and for detecting dangerous substances in security systems. This spectrometer is based on superconducting integrated receiver (SIR) developed in recent 15 years by our team in collaboration with Netherlands Institute for Space Research [4-5]. The SIR was built up in scope of TELIS project for monitoring the atmospheric compounds (ClO, HCl, H₂O isotopes, NH, O₃, BrO etc) at high altitude in the frequency range 490 - 630 GHz and demonstrated perfect operation at extreme flight conditions. While operating at altitude 20-35 km the absorption pass length (effective path of the beam at which the radiation is absorbed) reaches tens of kilometers, but in laboratory conditions we are strongly limited by dimensions of gas cell. The main purpose of



current work is to develop the compact laboratory spectrometer which can provide high sensitivity of detecting important gases.

2. Experimental setup & technique

The spectrometer under development uses the SIR as the receiver in 450-700 GHz range. The SIR is heterodyne receiver that utilizes extremely high non-linearity of tunnel Josephson junction “superconductor-insulator-superconductor” (SIS). The SIS-mixer is based on high-quality trilayer Nb/AlO_x/Nb or Nb/AlN/NbN with critical current density 5-10 kA/cm² fabricated on Si substrate and integrated with planar quasioptical dipole antenna, its dimensions are about 0,8 x 0,8 μm². The superconducting flux-flow oscillator (FFO) is long Josephson junction is used as the on-chip local oscillator, SIR intermediate frequency (IF) range is 4-8 GHz. For FFO stabilization an additional harmonic SIS-mixer (HM) is used in the feedback phase-lock loop system. All the elements needed for heterodyne detection are located on the 4x4 mm² microchip placed on the flat back surface of the silicon lens. The SIR block diagram is shown in figure 1. The concept, fabrication and experimental technique of SIR with more details are discussed in [4-6], the most important results of TELIS flight missions are presented in [6-8]. As for the cooling system for SIR microchip and other elements operating at low temperatures, two different systems are used. The first one is vacuum cryostat filled with liquid helium (helium volume is about 3.5 l) having the additional compartment for liquid nitrogen (about 2.5 l). Another option is to use closed cycle cooling system based on cryogenic refrigerator, which is more complicated but consumes only power supply and needs no liquid helium. In both cases, operating SIR temperature is about 4.2-5 K. By now the best double sideband noise temperature of receiver as low as 120 K is obtained, operating frequency range is from 450 to 700 GHz with continuous frequency tuning of the phase-locked local oscillator having free-running linewidth between 0.3 and 5 MHz, the spectral resolution is better than 0.5 MHz.

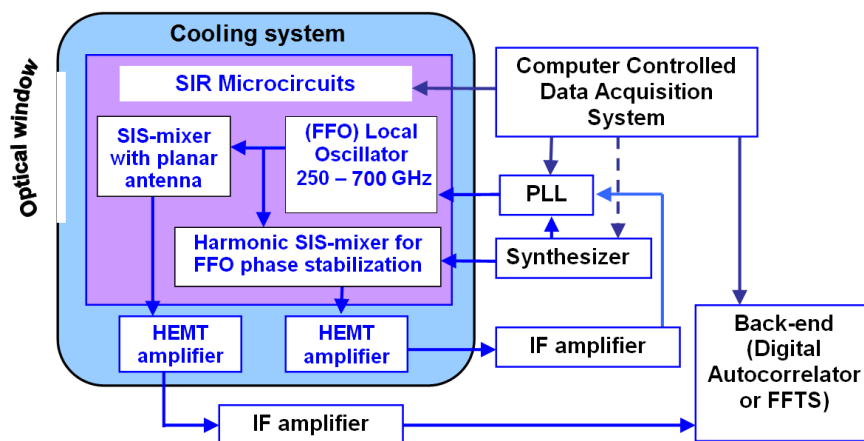


Figure 1. Block diagram of the superconducting integrated receiver (SIR) [6].

The simplified diagram of spectrometer setup for measurements of absorption lines is shown in figure 2. To measure the absorption spectral lines of gas samples by SIR we use the backward-wave oscillator (BWO) with a frequency multiplier and gas cell. The BWO has the operating frequency range 120-160 GHz, it was developed and fabricated in Institute for Physics of Microstructures. The BWO output signal is multiplied by quantum semiconductor superlattice (QSSL) multiplier by factor of 3-5 forming the output signal in the range 0.36-0.8 THz [9] passing then through the gas cell with the gas sample. The gas cell is a 60 cm length tube made of stainless steel, the diameter of cell of about 10 cm is large enough to reduce significantly the influence of absorbed molecules to the side surface of the cell on the results of measurement. The cell has the windows made of polyethylene

2 mm thick. To set and control the gas pressure in the range from atmospheric pressure to $\sim 10^{-6}$ mbar the turbo-molecular pump and the pressure gauge are used.

The measurement technique of the absorption lines is as described below. The output operating frequency of BWO with QSSL multiplier is tuned to the precise frequency (say, f_0) of the specified gas in THz range, the SIR detecting frequency is also tuned to the same frequency f_0 according to $f_{\text{FFO}} \pm f_{\text{IF}}$ (plus or minus depends on choice of upper or lower sideband operation). Then the multiplied frequency of BWO is slightly changed in the range $f_0 \pm \Delta$, where Δ should be deliberately larger than the width of absorption line. If necessary, the SIR operating frequency is also tuned in a simple way to search the spectrum or/and record the spectrum wider than IF range. Thus, using described technique absorption line is directly seen on the screen of spectrum analyzer and recorded by computer.

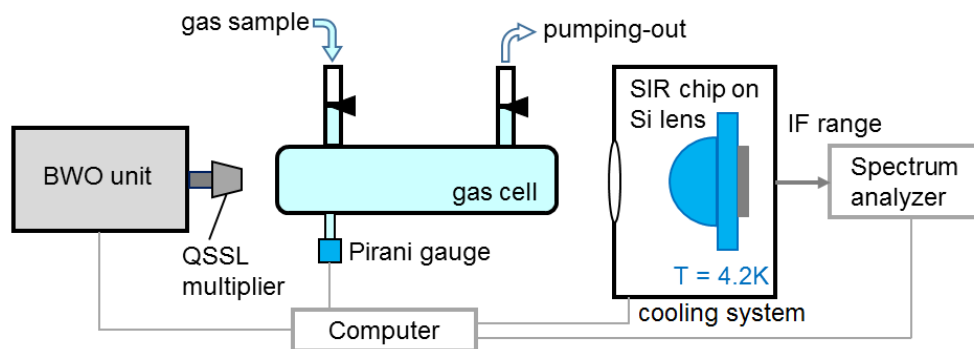
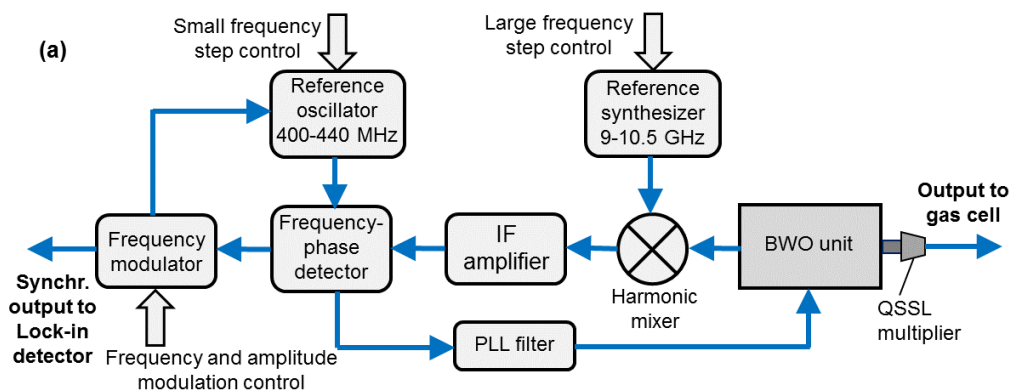


Figure 2. Experimental setup scheme for gas absorption spectra research at 450-700 GHz by SIR. Cooling setup based on liquid helium cryostat or closed cycle system can be used.

Another measurement technique that allows to improve significantly signal-to-noise ratio is using the synchronous detection. This methodic utilizes the frequency modulation of BWO output signal by frequency 0.2-1.5 MHz (with frequency deviation up to 5 MHz) and detecting the SIR output signal in a narrower range of 0.1-0.8 GHz by another back-end detector setup. In figure 3a the scheme of frequency modulation of BWO is shown, the signal from frequency modulator then comes as a reference signal to lock-in amplifier which is connected to SIR output. In figure 3b the scheme of signal storage and recording is presented.



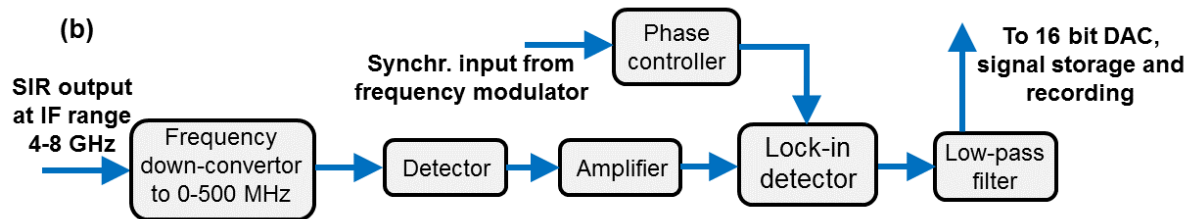


Figure 3. (a) Block diagram of BWO signal frequency and amplitude modulation. (b) Block diagram of detecting the output SIR signal by lock-in detector and its storage and recording.

3. First results and discussion

For the first testing measurements, the most common and simple in preparation and handling gases were chosen: ammonia NH_3 and water vapour. Their strong absorption lines in SIR operating range are at the frequencies 572.5 GHz and 556.9 GHz accordingly. The measured NH_3 absorption lines at different pressures with using spectrum analyzer as the back-end and without BWO frequency/amplitude modulation are presented in figure 4. It is clearly visible by our setup that the intensity of absorption depends on the gas pressure; the pressure limit for NH_3 to distinguish its presence turned out to be about 10^{-3} mbar. The absorption line of water vapour is stronger, hence the pressure limit for water in our measurements turned out to be better than 10^{-5} mbar. We assume this resolution limit can be considerably improved by using multi-pass gas cell and measuring technique with frequency modulation and lock-in detector as described in figure 3. Nevertheless, these results demonstrate the applicability of such technique and show the spectrometer under development to be an actual device for spectral analysis of gases.

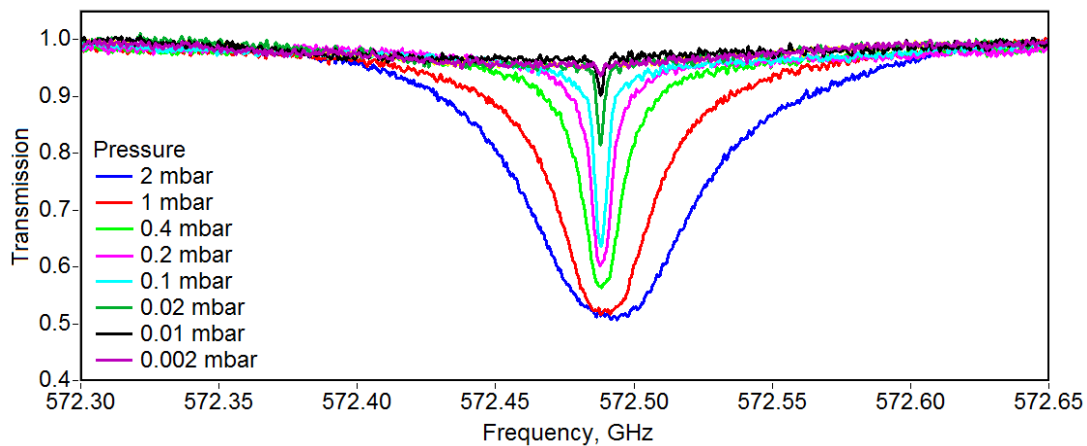


Figure 4. Absorption lines of ammonia NH_3 measured by SIR. Transmission level is calculated from the SIR output IF signal power measured by spectrum analyzer as back-end.

4. Conclusion

The new spectrometer with high sensitivity for gas analysis in 450-700 GHz range is being developed and presented in this work. The spectrometer is based on superconducting receiver with a noise temperature as low as 120 K and having SIS-mixer made of Nb/AlOx/Nb or Nb/AlN/NbN junctions. The introduced technique and the spectrometer under development can have many important practical applications. We concentrate primarily on medicine application, such as spectral non-invasive analysis of exhaled air. By detecting marker-substances this diagnostics can efficiently determine many different human diseases such as oncology, asthma, oxidative stress, schizophrenia, radiation injury, diseases of blood, respiratory tract, gastro-enteric tract, liver, kidney etc at different stages. Another

field of great importance nowadays that can require such devices is development of new security systems for detecting different explosive or poisonous substances, or, for instance, leakage of some gases in high-technology processes or on manufacture. Finally, such a device can be used for monitoring of atmosphere and ecological processed, climate changed and others.

Acknowledgments

The work is supported by Ministry of Education and Science of the Russian Federation (project No. 14.607.21.0100, ID RFMEFI60714X0100).

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