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METALS AND SUPERCONDUCTORS

Development of the Physical Principles of the Design and Implementation of a 500–700 GHz Spectrometer with a Superconducting Integrated Receiver

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Abstract—A spectrometer based on the effect of freely decaying polarization in the frequency range 500–700 GHz has been designed. Radiation sources are harmonics from a quantum semiconductor superlattice frequency multiplier. The receiving system of this spectrometer is constructed using a superconducting integrated receiver based on a superconductor–insulator–superconductor mixer and a flux–flow oscillator operating as a heterodyne oscillator. The spectrometer has been used to measure absorption lines of NH_3 in a sample of expired air (572 GHz).

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The terahertz (THz) and sub-terahertz (sub-THz) frequency ranges are very attractive for the use in many spectroscopic and, primarily, precise analytical investigations, because it is in these frequency ranges that the highest intensity absorption lines of many important materials have been observed. In this respect, the implementation of a high-sensitivity spectrometer that is suitable for the control over high-tech processes, the use in medical diagnostics, and the design of safety related systems seems to be a very important and topical problem.

Nowadays, there exists only one commercial spectrometer of this class. The Microtech Instruments Inc. (the United States) has developed a spectrometer operating in the sub-THz frequency range and employing a frequency-multiplied backward-wave oscillator, which covers the range from 100 to 1500 GHz. In this instrument, a standard pyroelectric detector is used as the receiving system. The spectrometer operates with a spectral resolution of 1–10 MHz and a sensitivity of the order of 5×10^{-7}. These characteristics are sufficient for solving a number of spectroscopic problems; however, there exist applications that require a higher frequency resolution and a higher sensitivity of the analysis. First and foremost, the case in point is the analysis of multicomponent gas mixtures in which the concentration of individual components can be at the level of ppb or even ppt. As an example of such problems, we should note the determination of impurities in high-purity materials, detection of toxic gases in the ambient air, monitoring of the processes occurring in chemical reactors, etc.

Among the existing methods of gas analysis, the best approximation to the theoretical threshold of sensitivity and the good frequency resolution limited only by the Doppler effect are provided by nonstationary microwave spectroscopy based on the effect of coherent spontaneous radiation [1–3]. The applicability of these methods in the sub-THz frequency range has become possible owing to the use of quantum semiconductor superlattice (QSSL) mixers and multipliers [4]. It has been demonstrated that these structures are more effective for the frequency conversion [4], because, in this case, as compared to Schottk diodes, the inertia of an electron transit through the active region and the parasitic capacitance become smaller, which makes it possible to increase the boundary operating frequency of the diode. Moreover, the quantum semiconductor superlattice has a current–voltage characteristic with a negative differential conductivity, which is retained up to frequencies above 1 THz. The QSSL mixers were used in the design of a new family of frequency synthesizers operating in the ranges 667–857, 789–968, and 882–1100 GHz [5], as well as a solid-state harmonic generator based on a Gunn gen-

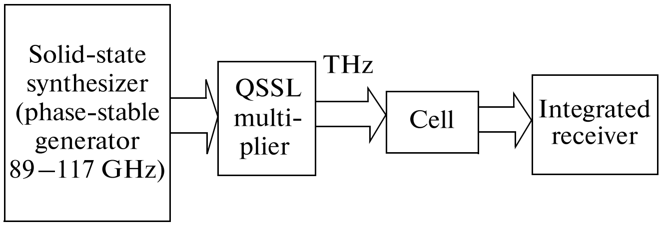

Fig. 1. Schematic diagram of the terahertz frequency spectrometer with a superconducting integrated receiver.

erator operating at frequencies up to 8.1 THz. Compared to the existing sub-THz sources produced by Microtech Instruments Inc., the harmonic generators are compact in form and simple in operation and have a longer service life.

Among the existing receiving systems, which operate in the sub-THz range, the most suitable version for the use in the design of a high-sensitivity spectrometer is the receiver based on a superconductor–insulator–superconductor (SIS) mixer and a flux–flow oscillator (FFO) employed as a heterodyne oscillator [6–9]. The sensitivity of this receiver, which is close to the quantum limit, is several orders of magnitude higher than the sensitivity of the existing receivers (piezoelectric sensors, thermocouples, Schottky diode detectors). The superconducting integrated receiver (SIR) also has a number of advantages, such as the compact form, the wide range of FFO frequency tuning, and the low energy consumption [10, 11].

At present, the frequency range of the majority of heterodyne receivers is limited by the heterodyne frequency tuning with typical values of 10–15% for a circuit of solid-state multipliers [12]. The bandwidth in the SIR is determined by a tunable structure of the SIS mixer and the matching circuit between the SIS and the FFO. A bandwidth up to 30–40% can be achieved by the design integration of a pair of SIS mixers. Another potential advantage is the use of a lattice of SIR channels inside one cryostat, which can operate at the same frequency or at different frequencies of the heterodyne oscillator.

In this paper, we present a spectrometer with phase manipulation of the exposing radiation and a superconducting integrated receiver operating in the frequency range 500–700 GHz. The physical principle of the spectrometer operation is as follows: the interaction of frequency-modulated radiation with resonantly absorbing molecules results in a periodic process of induction and decay of macroscopic polarization of the molecules [1]. A signal reemitted by the molecules lags behind the emission signal in phase, and this effect is used to receive a desired signal.

The simplified schematic diagram of the spectrometer is presented in Fig. 1. A signal from a reference generator is multiplied with the use of solid-state devices, i.e., quantum semiconductor superlattices. In the proposed scheme, gas molecules interact with a resonant THz harmonic. The high-Q cell can be used to increase the power of the resonant mode and to suppress the other modes.

Radiation sources in the THz range are the harmonics (up to 54th harmonic in the vicinity of 8100 GHz) obtained by multiplying the frequency of a synthesizer based on a Gunn generator operating in the frequency modulation mode in the frequency range 100–120 GHz with the use of a QSSL multiplier.

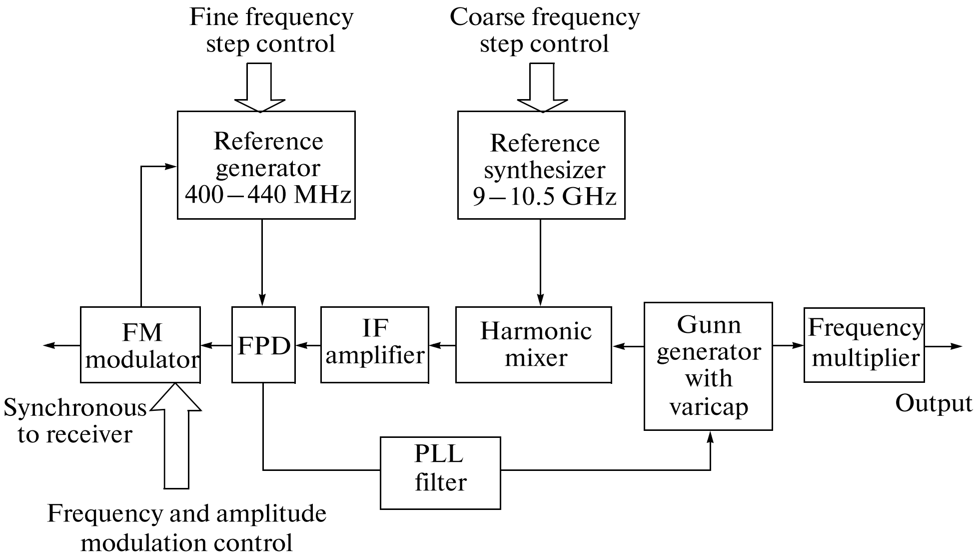

Fig. 2. Schematic diagram of the solid-state frequency synthesizer.

The schematic diagram of the solid-state frequency synthesizer with radiation frequency modulation is presented in Fig. 2. The harmonic of a reference coarse-frequency-step synthesizer operating in the frequency range 9.0–10.5 is mixed on a harmonic mixer with a part of the separated power of the output signal from the Gunn generator. The intermediate-frequency (IF) signal in the range 400–440 MHz is amplified and fed to a frequency-phase detector (FPD) in the phase-locked loop (PLL) system. The reference synthesizer for FPD is the fine-frequency-step synthesizer operating in the range 400–440 MHz with a minimum frequency step of 10 kHz. The signal from the FPD output passes through a PLL filter and is fed to the frequency control input of the Gunn generator, thus closing the PLL system.

The frequency modulator sets the frequency and deviation of the reference IF generator, which then are transferred to the output signal of the Gunn generator. The PLL band is chosen so as to transfer the frequency modulation from the IF channel to the output signal without a distortion. All parameters of the solid-state synthesizer, namely, the radiation frequency and modulation parameters, are set from a computer through a microcontroller.

The receiving system of the spectrometer is designed using a superconducting integrated receiver based on a SIS mixer and an FFO operating as a heterodyne oscillator. The schematic diagram of the SIR microchip (4.0 × 4.0 × 0.5 mm in size) designed at the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences (Moscow, Russia) is presented in Fig. 3. The frequency resolution of the receiver (along with the noise temperature and directional pattern) is one of the main parameters of the spectrometer. In order to obtained the required frequency resolution, the superconducting heterodyne oscillator of the integrated receiver should be synchronized to the reference synthesizer. For these requirements to be satisfied, we developed the concept of an integrated receiver with the cryogenic PLL system of the heterodyne oscillator [13]. According to this concept, a signal from the superconducting heterodyne oscillator is distributed between two SIS mixers, one of which is used as a receiving quasiparticle element and the other operates as a harmonic mixer in the PLL system.

The integrated receiver operates under the following conditions: the frequency range is 500–700 GHz, the noise temperature is lower than 200 K, the IF band is 4–8 GHz, the directional pattern with side lobes is at the level of less than –17 dB, and the spectral resolution is 1 MHz. Compared to the existing systems with close parameters, the proposed spectrometer is characterized by a wider range of input frequencies, smaller dimensions, and a lower energy consumption.

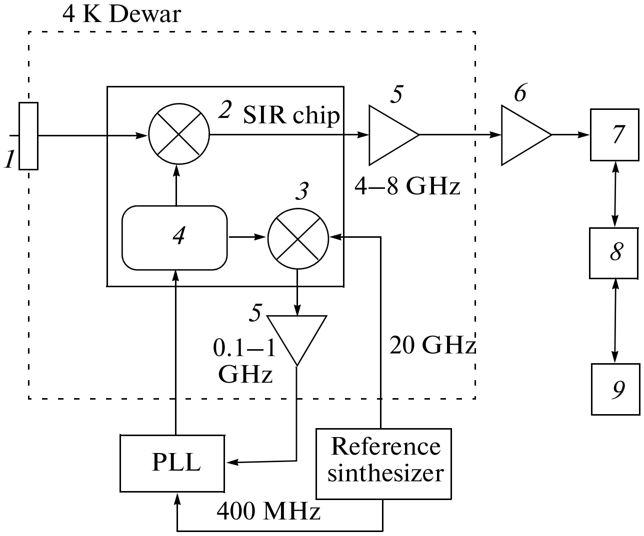

Fig. 3. Schematic diagram of the superconducting integrated receiver (SIR) chip in a Dewar vessel (indicated by the dotted line) at a temperature of 4 K: (*1*) optical input, 500–700 GHz; (*2*) superconductor–insulator–superconductor (SIS) mixer; (*3*) harmonic mixer (HM); (*4*) flux-flow oscillator (FFO) operating as a heterodyne oscillator, 500–700 GHz; (*5*) high electron mobility transistor (HEMT) amplifiers; (*6*) amplifier; (*7*) intermediate frequency (IF) processor and digital-to-analog converter (DAC); (*8*) control and data processing system; and (*9*) FFO, SIS, and HM control system.

Figure 4 shows the schematic diagram of the IF processor and the data collection system with a mixer in which the output signal in the range 4–8 GHz is mixed with a reference signal at 3.6 GHz. The IF signal passes through a 0.4–0.8 GHz bandpass filter and is fed to a Schottky-barrier diode detector.

The separated modulation signal is amplified by a video-frequency amplifier and fed to the input of the phase-locked detector. For this detector, the reference signal is the phase-controlled signal from the frequency modulator.

The phase control makes it possible to separate the largest amplitude of the desired signal and to reduce interfering signals due to both the interference in the THz channel and irregularities in the IF channels.

After passing the low-pass filter, the desired signal is digitized using a 16-bit digital-to-analog converter (DAC), where the preliminary storage of signals takes place. The further storage is performed with software in the computer.

The performance of the spectrometer was tested in measurements of absorption spectra of a number of molecules. In particular, measurements were performed in samples of expired air at a frequency of the absorption line of ammonia (572 GHz), because, spectroscopically, the expired air represents a multicomponent gas mixture. The cell had the form of a glass tube 10 cm in diameter and 60 cm long with optically transparent flat windows at the ends. The large

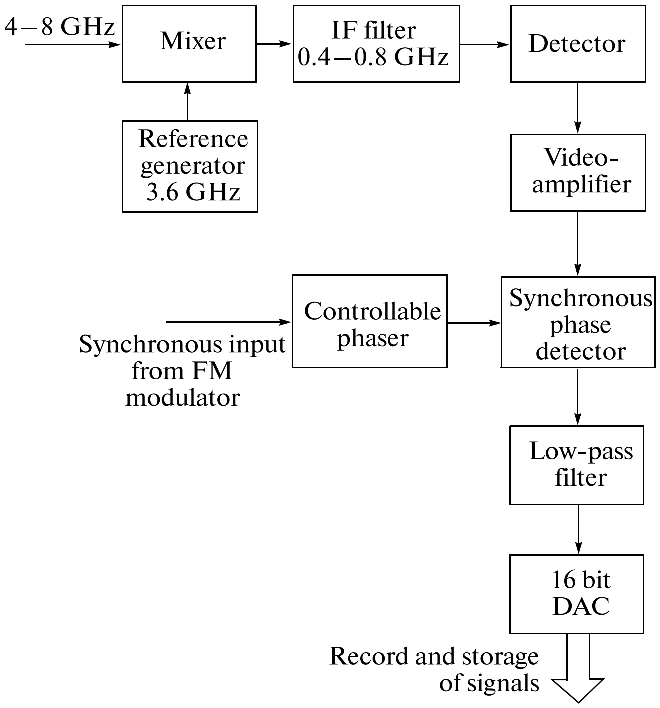

Fig. 4. Schematic diagram of the IF processor and data collection system.

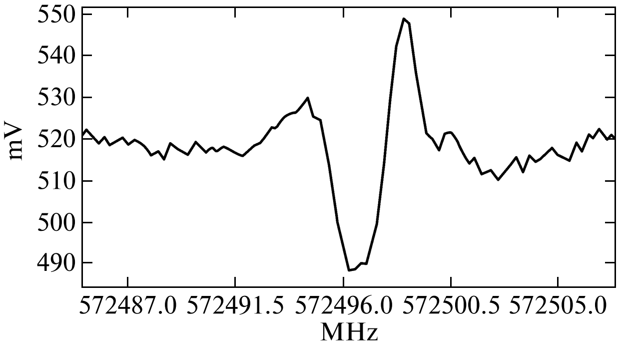

Fig. 5. Writing of the absorption line of NH_3 at a frequency of 572 GHz in the sample of expired air.

diameter of the cell made it possible to considerably decrease the influence exerted on the overall result of the measurements by gas molecules deposited onto the walls of the cell. The diameter of the radiation beam passing through the central axis of the cell did not exceed 3 cm. The products of gas exchange with the cell walls did not penetrate into this beam. The measurements were carried out in the regime of continuous circulation of the gas under investigation at a constant pressure of ~3×10^{-2} Torr attained in the cell. The calibration was performed using a 1% ammonia solution, which was additionally diluted in a 1/100 proportion. As a result, the gas concentration in the calibrated sample was equal to 10^{-4} mole fractions. For a more accurate calibration, it is necessary to prepare precision calibration mixtures and to check them using other methods. Figure 5 shows the writing of the absorption line of NH_3 in the sample of expired air. According to the above calibration, the NH_3 concentration in the expired air can be measured to a certain accuracy with the aim of subsequent medical diagnostics. Thus, the performed test measurements of rotational spectra of a number of basic molecules have confirmed the high sensitivity of the instrument (no worse than 1 ppb) with a spectral resolution limited only by the Doppler effect.

In conclusion, we note that, in this paper, the possibility of designing a laboratory-purpose sub-THz spectrometer based on a harmonic generator and a superconducting integrated receiver with a quantum sensitivity has been demonstrated for the first time. The characteristics of this spectrometer satisfy the requirements of the precision gas analysis.

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