Superconducting Integrated Terahertz Spectrometers

Valery P. Koshelets, *Member, IEEE*, Pavel N. Dmitriev, Michael I. Faley, Lyudmila V. Filippenko,
Konstantin V. Kalashnikov, Nickolay V. Kinev, Oleg S. Kiselev, Anton A. Artanov,
Kirill I. Rudakov, Arno de Lange, G. de Lange, Vladimir L. Vaks, M. Y. Li, and Huabing Wang

***Abstract*—A superconducting integrated receiver (SIR) comprises all of the elements needed for heterodyne detection on a single chip. Light weight and low power consumption combined with nearly quantum-limited sensitivity and a wide tuning range of the superconducting local oscillator make the SIR a perfect candidate for many practical applications. For the first time, we demonstrated the capabilities of the SIR technology for remote operation under harsh environmental conditions and for heterodyne spectroscopy at atmospheric limb sounding on board a high-altitude balloon. Recently, the SIR was successfully implemented for the first spectral measurements of THz radiation emitted from intrinsic Josephson junction stacks (BSCCO mesa) at frequencies up to 750 GHz; linewidth below 10 MHz has been recorded in the high bias regime. The phase-locked SIR has been used for the locking of the BSCCO oscillator under the test. To extend the operation range of the SIR well above 1 THz, a new technique for fabrication of high-quality SIS tunnel junctions with gap voltage V_g up to 5.3 mV has been developed. Integration of a superconducting high-harmonic phase detector with a cryogenic oscillator opens a possibility for efficient phase locking of the sources with free-running linewidth up to 30 MHz that is important both for BSCCO mesa and NbN/MgO/NbN oscillators.**

***Index Terms*—Oscillators and spectrometers, phase locking, superconducting integrated circuits, terahertz receivers, thin-film circuits, tunnel junctions.**

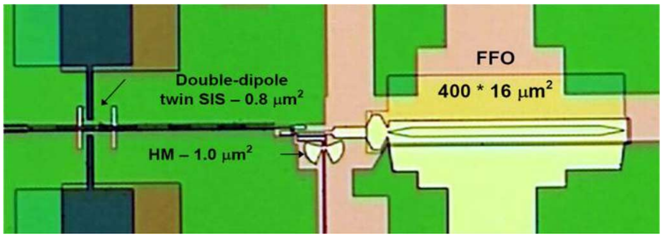

Fig. 1. Central part of the SIR chip with antenna, twin SIS-mixer, and harmonic mixer for FFO phase-locking [4].

I. Introduction

A SUPERCONDUCTING integrated receiver (SIR) [1]–[4] comprises on a single chip a low-noise SIS mixer with quasioptical antenna, a flux-flow oscillator (FFO) acting as a local oscillator (LO), and a second SIS harmonic mixer (HM) for the FFO phase locking (see Fig. 1). The concept of the SIR looks very attractive for many practical applications due to its compactness and wide tuning range of the FFO; a bandwidth of up to 35% has been achieved with a twin-junction SIS mixer design. Recently, the frequency range of most practical heterodyne receivers was limited by the tunability of the local oscillator; nowadays, commercially available multipliers cover the band up to 40% of the center frequency,[1] and the best SIS receivers[2] offer the bandwidth 15%–30%. All components of the SIR microcircuits are fabricated out of high-quality Nb-based tri-layer on Si substrate, placed on the flat back surface of the silicon lens.

Continuous tuning of the phase-locked local oscillator has been realized at any frequency in the range 300–750 GHz [3], [4]. The output power of the FFO is sufficient to pump the matched SIS mixer in a wide frequency range and can be electronically adjusted. The FFO free-running linewidth has been measured between 0.3 and 5 MHz, resulting in the spectral ratio of the phase-locked FFO above 70% over the range. As a result of receiver optimization, the DSB noise temperature was measured below 100 K, which is about $4hf/k_B$; the spectral resolution is well below 1 MHz [3], [4].

All of these achievements enabled the development of a 450–650-GHz integrated receiver for the atmospheric-research instrument TELIS (TErahertz and submillimeter LImb Sounder)—the balloon-borne instrument for the detection of spectral emission lines of stratospheric trace gases that have their rotational transitions at THz frequencies [4], [5], [6]. Diurnal cycle of ClO has been observed; the BrO line with a level of only 0.3 K was isolated and clearly detected. Some recently obtained TELIS results are presented in Section II. Capability of the SIR for high-resolution spectroscopy has been successfully proven also in laboratory environment by gas cell

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V. P. Koshelets, P. N. Dmitriev, L. V. Filippenko, N. V. Kinev, and O. S. Kiselev are with the Kotel'nikov Institute of Radio Engineering and Electronics, Moscow 125009, Russia (e-mail: valery@hitech.cplire.ru).

M. I. Faley is with the Kotel'nikov Institute of Radio Engineering and Electronics, Moscow 125009, Russia, and also with PGI-5 (Microstructure Research), Forschungszentrum Jülich GmbH, D-52428 Jülich, Germany.

K. V. Kalashnikov, A. A. Artanov, and K. I. Rudakov are with the Kotel'nikov Institute of Radio Engineering and Electronics, Moscow 125009, Russia, and also with the Moscow Institute for Physics and Technology (State University), Dolgoprudny 141700, Russia.

A. de Lange and G. de Lange are with the SRON Netherlands Institute for Space Research, 3584 CA Utrecht, The Netherlands.

V. L. Vaks is with the Institute for Physics of Microstructure and Lobachevsky State University, Nizhny Novgorod 603950, Russia.

M. Y. Li and H. B. Wang are with the Research Institute of Superconductor Electronics, Nanjing University, Nanjing 210093, China, and also with the National Institute for Materials Science, Tsukuba 3050047, Japan.

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[1] [Online]. Available: http://vadiodes.com/index.php/en/products/full-band-multipliers-wr-series

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measurements. The possibility to use SIR devices for the medical analysis of exhaled air has been demonstrated [3]. Many medically relevant gases have spectral lines in the sub-terahertz range [7] and can be detected by a SIR-based spectrometer.

Recently, the SIR was successfully implemented for the first spectral measurements of THz radiation emitted from intrinsic Josephson junction stacks (BSCCO mesa); some of the lately obtained spectral data as well as the first result of BSCCO mesa phase-locking to external reference is presented in Section III. A possibility to extend an operation frequency of the SIR beyond 1 THz will be discussed in Section IV.

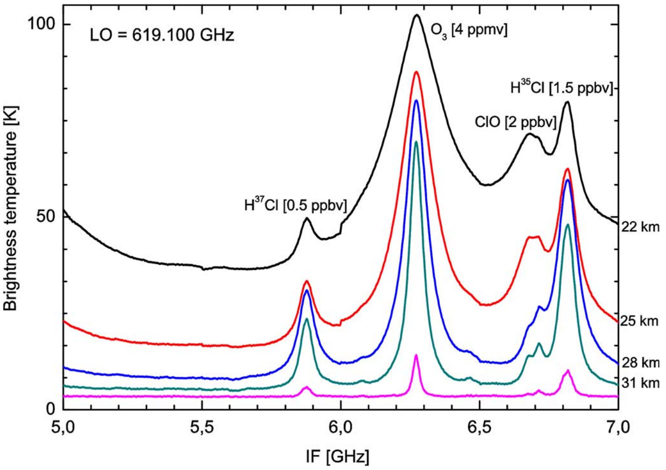

Fig. 2. Spectra of two HCl isotopes, ozone and ClO; LO frequency is 619.1 GHz. Spectra for tangent heights 22–31 km and up-looking 6° measured in Kiruna, 2010, are presented in the graph. Corresponding estimated concentrations of observed gases are shown.

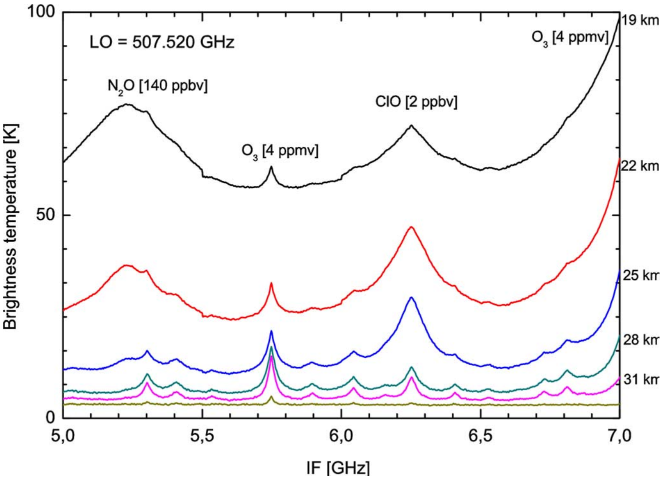

Fig. 3. Spectra of ClO, ozone and N_2O; LO frequency is 507.52 GHz. Spectra for tangent heights 19–31 km and up-looking 6° measured in Kiruna, 2010, are presented in the graph.

II. Recent TELIS Results

The TELIS instrument is a three-channel balloon-borne heterodyne spectrometer [8] developed by collaboration of four institutes: the Deutsches Zentrum für Luft- und Raumfahrt (DLR), Germany, the Rutherford Appleton Laboratories (RAL), United Kingdom, and the SRON—Netherlands Institute for Space Research, The Netherlands (in close collaboration with the Kotel'nikov Institute of Radio Engineering and Electronics, IREE, Moscow). The TELIS is a compact, lightweight instrument capable of providing information about spectral lines presented in both sub-THz (SIR channel by SRON-IREE) and THz (1.8 THz HEB-mixer developed by DLR) spectral regions.

The TELIS shares the balloon platform with the Fourier transform spectrometer MIPAS-B [9], developed by the Institute of Meteorology and Climate research of the University of Karlsruhe, Germany (IMK) and is operated in the mid-infrared range (680 to 2400 cm^{-1}). Both instruments observe simultaneously the same air mass, and together they can provide information about many atmospheric trace gases measured simultaneously in several spectral regions, which provides an opportunity for cross-check results of different instruments. The instrument has been proven to be stable against the strong atmospheric temperature variations during the ascent (with ambient temperatures as low as -90 °C).

TELIS instrument had four successful flights: three in Kiruna, Sweden (2009, 2010, and 2011) and one in Timmins, Canada (2014). During all of those flights, the shortest of which lasted 10 h on float, were measured thousands of limb spectra. Examples of recently elaborated spectra measured by the SIR-TELIS channel at different LO frequencies are presented in Figs. 2 and 3.

The flights in North Sweden focused on catalytic ozone loss by halogens in the Arctic region, similar to processes causing the infamous ozone-hole over the Antarctic. The spectra depicted in Figs. 2 and 3 cover some core molecules to address these processes, namely ozone itself and Cl-bearing species HCl and ClO. ClO is the main form of active chlorine causing the catalytic destruction of ozone. HCl, on the other hand, is a so-called reservoir species as it is mostly inert in ozone chemistry. The ozone destruction depends on the ClO concentration which strongly depends on altitude. Limb sounding provides a tool to gain insight in the vertical distribution of these concentrations. A single recording of a spectrum contains mostly information of molecules at tangent point, which is the lowest point in the atmosphere of the light path. By combining several spectra recorded at different tangent heights, a vertical profile can be constructed. The integration time of a single spectrum is 1.5 s and a typical limb scan contains typically 10–20 different recordings, covering tangent heights between 10 and 35 km, for a total measurement time, including calibration measurements, close to 1 min. In the figures, only a few spectra from a limb scan are shown to prevent cluttering.

For the results presented in Figs. 2 and 3 limb scans data were averaged 10 and 23 times correspondingly. It gives the total averaging time for one tangent height about 16 s (Fig. 2) and 35 s (Fig. 3). To provide the same signal-to-nose ratio (SNR) for the semiconductor-based receivers, which have noise temperature not lower than 1800 K [10] in the same frequency range, measurement time should be increased for two orders of magnitude at least. The wideband coverage of the FFO gives the advantage that huge amount of molecules which absorption lines lay in the FFO tuning range, can be measured during one campaign. The TELIS-SIR channel has been characterized in eight micro-windows covering the FFO frequency range from 495.04 GHz (for

$H_2{}^{18}O$) to 619.10 GHz (for HCl, ClO and HOCl); in combination with short integration time, it gives an opportunity to provide vertical profiles for many molecules for almost the same air mass.

The final product of those measurements is presented in several papers concerning atmospheric chemistry [6], [11]–[13]. Analysis of all of the flight data is an ongoing process, where post-flight characterization of the SIR (for example, precise laboratory measurement of the SIR sideband ratio) gave a new input to processing of the flight data with higher accuracy. Already analyzed data proved the SIR to be one of the most sensitive sub THz spectrometers allowing to measure concentration of trace gases lower than 1 ppbv.

III. MEASUREMENTS OF THZ RADIATION FROM BSCCO MESA AND ITS PHASE-LOCKING TO EXTERNAL REFERENCE

In recent years, coherent THz emission have been obtained from stacks of intrinsic Josephson junctions (IJJs) made of the high temperature superconductor $Bi_2Sr_2CaCu_2O_8$ (BSCCO). An IJJ is formed naturally in the BSCCO unit cell with the CuO_2 layers forming the superconducting electrodes and the BiO and SrO layers forming the barrier layer [14]. A 1-μm-thick crystal consists of about 670 IJJs. In 2007, it was reported that such stacks can emit coherent radiation at frequencies up to 0.85 THz with a directly detected power of about 10 nW [15]. Terahertz emission from intrinsic BSCCO stacks has been obtained both at a low bias (where the temperature distribution in the stack is almost homogeneous) and a high bias regime (where an over-heated part and a cold part of the sample coexist) [16], [17]. Application of the SIR has allowed to measure radiation emitted from intrinsic Josephson junction stacks in both regimes with spectral resolution better than 1 MHz for the first time [17]. While at low bias we found that linewidth is not smaller than 500 MHz, at high bias, emission linewidth turned out to be in the range 10–100 MHz. We attribute this to the hot spot acting as a synchronizing element; linewidth as narrow as 7 MHz has been recorded at high bias [see Fig. 4(a)].

Typical dependencies of the linewidth on the BSCCO frequency both in the low-bias and high-bias regimes that were measured by the SIR are presented in the Fig. 4(b). Important to note that the tuning of the BSCCO oscillator frequency is continuous over the range; that was confirmed by fine tuning of the SIR LO frequency. Actually for the presented data the lowest measured frequency of about 550 GHz was limited by the BSCCO mesa, while losses in the Nb interconnection lines of the SIR restrict the measurements at frequencies higher than 750 GHz.

Coherent emission above 1 THz by intrinsic Josephson BSCCO junction stacks with improved cooling has been demonstrated [18], [19]. Due to the variable size of the hot spot and the temperature rise caused by the self-heating, the emission frequency can be tuned over a wide range of up to 700 GHz [18]. So far, emitted by one device power up to 30 μW was obtained [20], [21]; recently, by synchronizing the emissions from a three-mesa array, power as high as 610 μW was reported [22]. These are very encouraging results, although for most practical application phase-locking of the cryogenic oscillator to a stable reference is required.

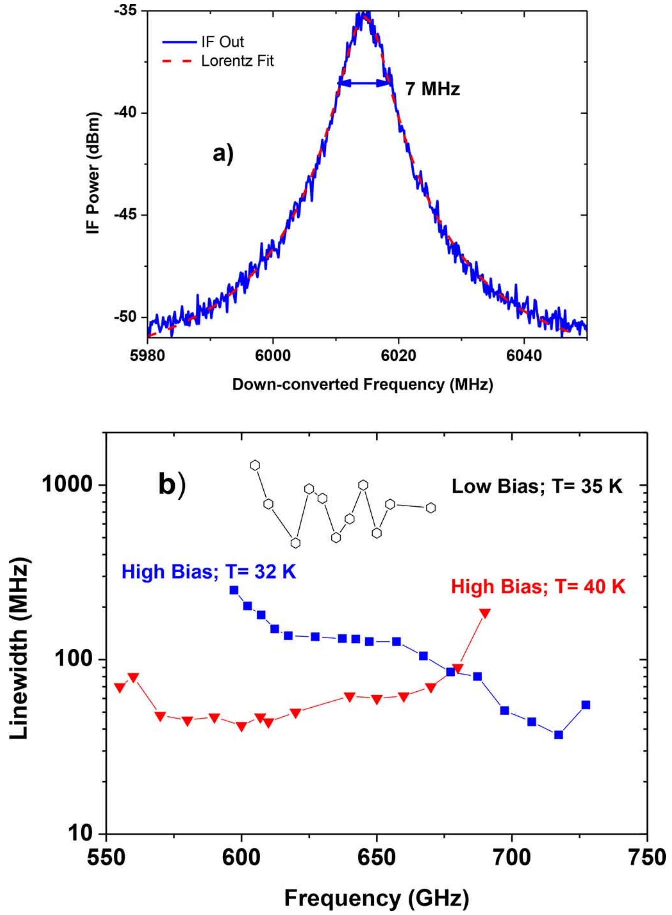

Fig. 4. (a) Down-converted spectrum of the BSCCO at 476 GHz in the high-bias regime; dashed line is a Lorenzian fit with full linewidth = 7 MHz. (b) Typical dependence of the BSCCO linewidth on the frequency (measured for the different sample)—solid (open) symbols are for high (low) bias regime.

To check a principal possibility of such locking we used the phase-locked SIR not only for detection of the BSCCO oscillator emission, but also for further locking of the oscillator under the test [23]. Block diagram of the experimental setup for phase locking of the BSCCO oscillator is presented in Fig. 5. The BSCCO oscillator signal initially down-converted by the SIR to the IF band 4–8 GHz was then down-converted one more time to a secondary IF band 0.1–0.9 GHz. The obtained IF signal is actually a convolution of the BSCCO oscillator signal and stable phase-locked SIR LO. This signal is applied to the PLL 2, where phase of the signal is compared with phase of the stable reference (f_{ref} = 400 MHz). Note that all reference sources used in the experiment (400 MHz, 6 GHz, and tunable 19–21 GHz) were internally synchronized to the common 10 MHz reference. The error signal is returned back to the BSCCO oscillator to control its phase via additional 5-Ω resistor mounted on the bias plate of the oscillator. It should be mentioned that both PLLs were equipped by additional frequency discriminator (FD), which compares signal with internal 400-MHz resonance tank; the FD error signal was applied to the oscillator in parallel with PLL signal and can be adjusted separately.

Results of the BSCCO oscillator frequency and phase locking are presented in Fig. 6. Linewidth of the BSCCO oscillator frequency locked at 563 GHz is 13.5 MHz [Fig. 6(a)]; about 10% of the oscillator power has been phase-locked. The ratio of the

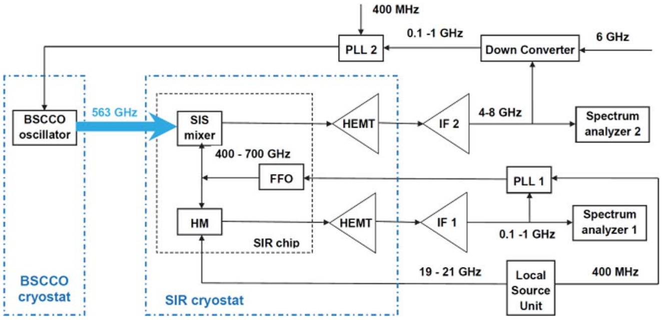

Fig. 5. Block diagram of the experimental setup for the BSCCO oscillator phase locking (see text for more details).

phase-locked power to the total power emitted by the oscillator is called a “spectral ratio” (SR); an obtained SR value of about 10% is a reasonably good result taking into account wide linewidth of the BSCCO oscillator and long length of the PL loop. An SNR of about 57 dB in a bandwidth of 1 Hz has been recorded [see Fig. 6(c)].

In the phase-locking experiments, we have found that, while higher power is favorable for phase-locking (especially because the PLL feed-back circuitry was not optimized for BSCCO oscillator phase-locking), the realization of the true voltage control oscillator with relatively small delay (compare to hot spot operation, where all voltage changes are rather slow) is also rather important. This situation is quite similar to the results obtained at the quantum cascade laser (QCL) phase-locking [24]. This first successful attempt to phase-lock a BSCCO oscillator to stable microwave reference opens prospects for numerous practical applications.

IV. TOWARDS THZ INTEGRATED RECEIVERS

For many years, tunnel junctions based on niobium nitride (NbN) have been attracting interest as an alternative to Nb junctions for high-frequency applications since NbN has large gap energy. There have been many reports on the development of NbN tunnel junctions using different tunnel barrier materials [25]–[28]. Initially, only NbN/MgO/NbN junctions have exhibited reasonably good quality so far, because both NbN and MgO have the same crystal structure with a lattice mismatch of less than 5%. Recently high-quality epitaxial NbN/AlN/NbN tunnel junctions with a wide range of current density have been demonstrated [29]. Although previous works have proven a possibility to produce high-quality all NbN tunnel junctions, we developed a new technique to fabricate NbN/MgO/NbN circuits. Our approach somehow resembles the “classical” technique proposed many years ago for the Nb/AlOx/Nb junctions [30], which are the basic building block for most devices of modern superconducting electronics. According to our approach very thin Mg layer (about 1.5 nm only) is dc sputtered on the NbN layer; the Mg is then oxidized in the O_2 plasma (similar to Al nitridization process used for fabrication of Nb/AlN/NbN junctions [31], [32]). Details of the fabrication process as well as results of the comprehensive measurements of obtained circuits will be presented elsewhere [33].

The transmission electron microscopy (TEM) image (see Fig. 7) shows the layer structure of the junction area of the

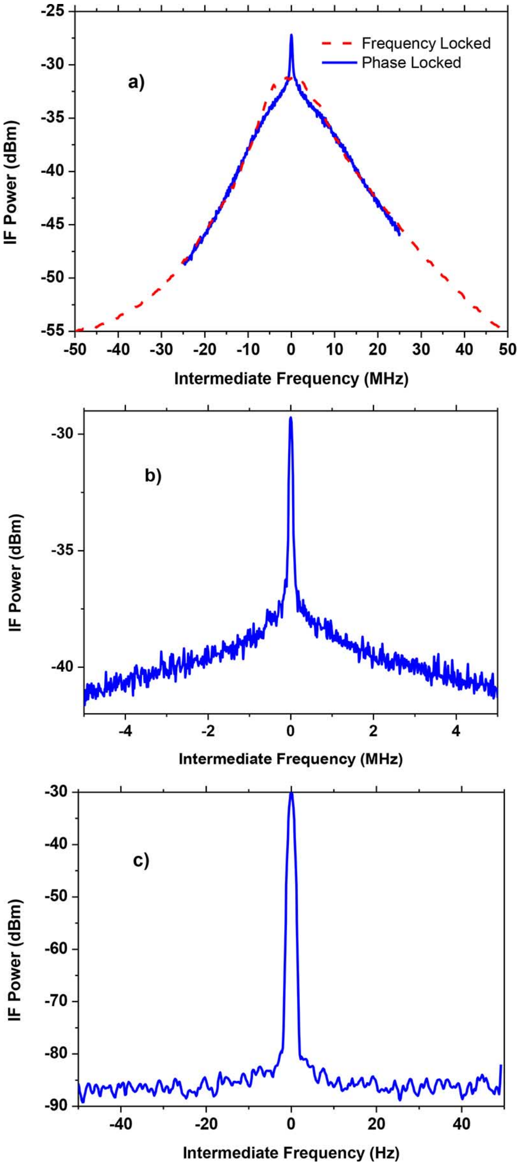

Fig. 6. Spectra of the BSCCO oscillator measured relative to the phase-locked FFO of the SIR: (a) Frequency (dashed) and phase locked (solid line). Span 100 MHz, resolution bandwidth $(\mathrm{RBW}) = 0.47$ MHz $\mathrm{Linewidth} = 13.5$ MHz. (b) Phase-locked, span 10 MHz, $\mathrm{RBW} = 9.1$ kHz. (c) Phase locked, span $=$ 100 Hz, $\mathrm{RBW} = 1$ Hz, SNR is 57 dB as measured in a bandwidth of 1 Hz.

sample. The MgO substrate, bottom and top NbN electrodes and MgO barrier are visible. Bottom electrode consists of the epitaxial 70 nm NbN monitor layer covered by 150-nm-thick NbN film, which is polycrystalline due to lift-off structuring of this and all subsequent layers. Top NbN electrode is polycrystalline and has thickness of 70 nm. High-resolution TEM image of 1.5-nm-thick MgO barrier layer is shown in the insert of the picture. It was observed that the orientation of crystal structure of NbN electrodes is maintained across the MgO barrier.

By using the developed technique it is possible to fabricate high-quality junctions with quasiparticle tunnel current density Jg in the range 0.05–80 kA/cm^2. The IVC of the NbN/MgO/NbN junction ($J_g = 2$ kA/cm^2) is shown in Fig. 8; the dependencies of the gap voltage Vg and quality factor Rj/Rn

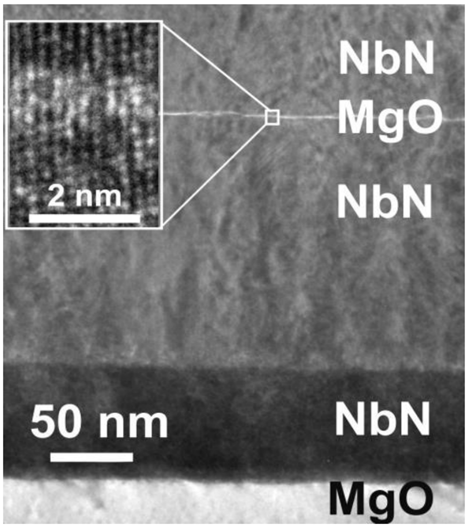

Fig. 7. Transmission electron microscopy image of the layer structure of the NbN–MgO- NbN junction.

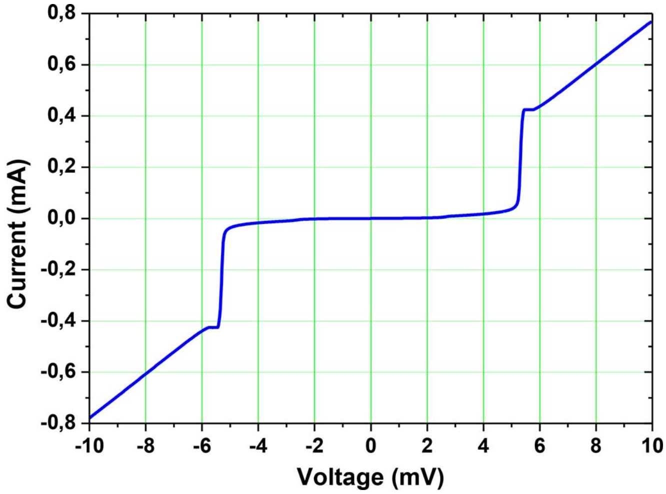

Fig. 8. IVC of the NbN/MgO/NbN junction (Ic is suppressed by magnetic field): $\mathrm{S} = 18\ \mu\mathrm{m}^2$; $\mathrm{Jg} = 2\ \mathrm{kA/cm}^2$ $\mathrm{Vg} = 5.3\ \mathrm{mV}$; $\mathrm{Rn} = 11.7\ \Omega; \mathrm{Rj}(2\ \mathrm{mV})/\mathrm{Rn} = 80, \mathrm{Rj}(4\ \mathrm{mV})/\mathrm{Rn} = 19.5$.

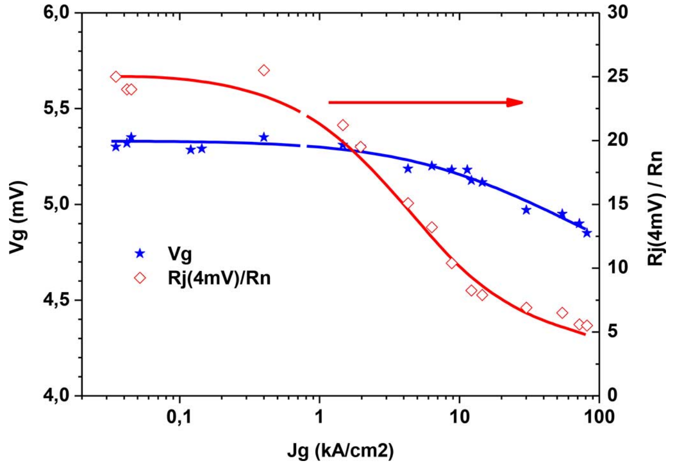

Fig. 9. Dependencies of the NbN-MgO-NbN junction' parameters on current density Jg. Experimental points are connected by lines as a guide for the eye.

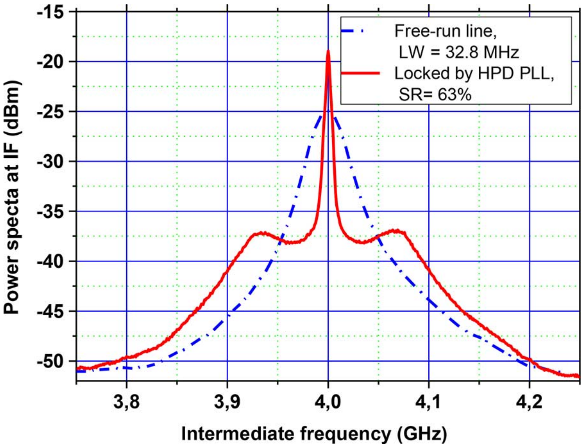

Fig. 10. Down-converted spectra of the FFO: frequency locked (dash-dotted line) and phase-locked by the HPD (solid line). The CPLL synchronizes up to 63% of emitted power for free-running FFO linewidth of about 33 MHz.

(ratio of the leakage resistance Rj and normal state resistance Rn) on the current density are presented in Fig. 9.

To summarize, new technique for fabrication of high-quality SIS tunnel junctions based on epitaxial NbN films with MgO barrier has been developed; the junctions with gap voltage $\mathrm{Vg} = 5.3\ \mathrm{mV}$ and quality barrier parameter $\mathrm{Rj}(4\ \mathrm{mV})/\mathrm{Rn} > 25$ have been fabricated. Such junction parameters are very promising for development of a SIR for frequencies well above 1 THz.

For efficient locking of Lorentzian lines a PLL system with a very wide regulation bandwidth (RegBW) is required (due to slow decrease of the noise level with offset from the carrier). The required RegBW depends on the oscillator linewidth [34]; to phase lock more than 90% of the emitted by the oscillator power (that is required for most applications in radio astronomy and high-precision spectroscopy) the RegBW should exceed 70 MHz for free-running linewidth of 10 MHz. To overcome the limitations of the traditional PLL, we have developed the Cryogenic Phase Locking Loop system (CPLL) [35]. Implementation of SIS junction both for down-conversion of oscillator frequency and generation of feedback signal allows us to place all PLL elements in close vicinity to the oscillator. In turn, this provides significant reduction of loop time delay (less than 4 ns) and extremely large RegBW (up to 70 MHz).

We called this novel element "high-harmonic phase detector (HPD)"; the basic principle of its operation is as follows: the FFO signal $\mathrm{f_{FFO}}$ (~600 GHz) is mixed by HPD with LO signal, which frequency $\mathrm{f_{LO}}$ (of about 20 GHz) is chosen to exactly satisfy the relation $\mathrm{n} * \mathrm{f_{LO}} = \mathrm{f_{FFO}}$. In this case the HPD generates low frequency output signal proportional to the phase difference between the FFO and the appropriate harmonic of the LO. This error signal is applied directly to the FFO control line through a low-pass filter. Since the CPLL system consists of only superconductive and low-consumption elements, it could be integrated on the single chip with locked oscillator. As it is shown in Fig. 10, the CPLL system could efficiently synchronize highly broad emission lines. So, the HPD approach is promising for NbN/MgO/NbN oscillator stabilization, which is expected to have large linewidth due to increased surface losses, as well as for the phase-locking of BSCCO oscillators by using high-Tc harmonic mixer [20].

V. CONCLUSION

Nowadays the SIR is probably the most functionally complex fully superconducting device that was already successfully implemented for practical applications such as Earth atmosphere monitoring. The SIR is very attractive for future airborne and space-borne missions as well as for analysis of exhaled air at medical survey and for security monitoring. New techniques for fabrication of high-quality SIS tunnel junctions with gap voltage $Vg > 5$ mV as well as approach for phase-locking of the cryogenic oscillators with linewidth up to 50 MHz have been developed to extend operation frequency of the SIR beyond 1 THz. The SIR was successfully implemented for the first spectral measurements of THz radiation emitted from intrinsic BSCCO Josephson junction stacks; linewidth as narrow as 7 MHz has been recorded in the high bias regime. The phase-locked SIR has been used for the locking of the oscillator under the test. That is the first, but very important step towards the development of fully high Tc phase-locked local oscillator.

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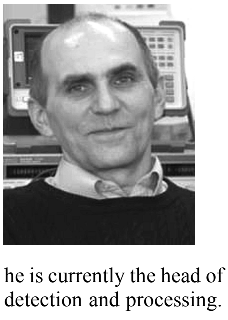


Valery P. Koshelets received the M.S. degree in physics from Lomonosov Moscow State University, Moscow, Russia, in 1973, and the Ph.D. degree in radio physics and Doctor of Sciences (Habilitation) degree in physical electronics from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, in 1978 and 1990, respectively.

Since 1993, he has been with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia, where he is currently the head of the Laboratory of superconducting devices for signal detection and processing.

Pavel N. Dmitriev received the M.S. degree from Lomonosov Moscow State University of Fine Chemical Technologies, Moscow, Russia, in 1993, and the Ph.D. degree from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, in 1993.

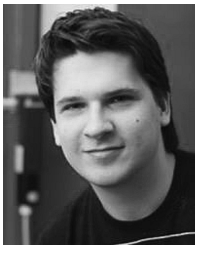
He is currently a Researcher with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia. His research interests include development and fabrication of superconducting circuits.

Michael I. Faley received the Diploma in physics (with honors) from the Moscow Institute of Physics and Technology, Moscow, Russia, in 1982, the Ph.D. degree in physical electronics (*summa cum laude*) from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, in 1987, and the Doctor of Science (Habilitation) degree in experimental physics (*summa cum laude*) in 2006. His dissertations were titled "Electron-electron interaction in disordered metal films" and "Thin film heterostructures of oxide superconductors and their application for superconducting quantum interference devices," respectively.

Since 1991, he has mainly been a Scientist with the Peter Grüberg Institute Forschungzentrum Jülich GmbH, Germany. His current research interests focus mainly on physics, technology, and prospective applications of high-T_c DC SQUIDs.

Lyudmila V. Filippenko received the M.S. degree from Saratov State University in 1986 and the Ph.D. degree from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia, in 2009.

She is currently a Senior Redearcher with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia. Her research interests include development and fabrication of superconducting devices.



Konstantin V. Kalashnikov received the M.S. degree from the Moscow Institute of Physics and Technology, Moscow, Russia, in 2011, and the Ph.D. degre in radiophysics from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia, in 2014. His dissertation was on experimental realization of new PLL system for superconducting integrating receivers.

He is currently a Scientist with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia, and an Assistant Teacher with the Moscow Institute of Physics and Technology. His research interests include superconducting electronics, such as heterodyne receivers, SIS mixers, and phase-locking systems.

Nickolay V. Kinev received the B.S. degree in applied mathematics and physics and M.S. degree in solid-state electronics and radio physics from the Moscow Institute of Physics and Technology, Moscow, Russia, in 2007 and 2009, respectively, and the Ph.D. degree from the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, in 2013. His dissertation was on the study and development of THz oscillators and receivers using superconducting devices.

He is currently a Researcher with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia. His research interests include superconducting receivers and oscillators for THz technologies and space applications.

Oleg S. Kiselev received the M.S. and Ph.D. degrees from the Moscow Institute of Physics and Technology, Moscow, Russia, in 2008 and 2011, respectively, both in experimental physics.

Since 2006, he has been a Researcher with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia, focusing mainly on balloon-borne instrument development. His main research interests include application of terahertz and superconducting instruments in lab spectroscopy and remote sensing.

Anton A. Artanov received the B.S. degree from the Moscow Institute of Physics and Technology, Moscow, Russia, in 2013, where he is currently working toward the M.S. degree.

Since 2012, he has been with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia. His research is mainly focused on SIS mixers for up- and down-conversion.

Kirill I. Rudakov received the B.S. and M.S. degrees in applied physics and mathematics from the Moscow Institute of Physics and Technology, Moscow, Russia, in 2013 and 2015, respectively.

He is currently a RF/HF Engineer with the Kotel'nikov Institute of Radio Engineering and Electronics of the Russian Academy of Sciences, Moscow, Russia. He took part in the ALPHA experiment at CERN and with the CHAMP+ experiment at SORN.

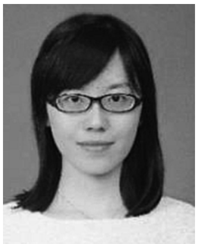
Arno de Lange received the M.Sc. and Ph.D. degrees in laser spectroscopy from the Free University of Amsterdam, Amsterdam, The Netherlands, in 1997 and 2002, respectively.

After two postdoctoral positions with the Free University of Amsterdam, Amsterdam, The Netherlands, and Imperial College London, London, U.K., both pertaining to ion-trapping, he joined SRON—Netherlands Institute of Space Research in 2005. There, he shifted his research focus to atmospheric sciences. Initially his interest was the balloon experiment TELIS, in which he was responsible for the data reduction of the SIR channel with a scientific focus on stratospheric processes in general and ozone depleting chemistry in particular. Currently, he is involved with satellite retrievals concerning greenhouse gases with a strong emphasis on algorithm development to further the quality of the retrieval products.

G. de Lange, photograph and biography not available at the time of publication.

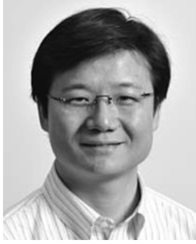
Vladimir L. Vaks was born in 1946. He received the B.S. and Ph.D. degrees from Nizhny Novgorod State University, Nizhny Novgorod, Russia, in 1970 and 2003, respectively.

He is currently a Head of the Terahertz Spectrometry Department, Institute for Physics of Microstructures, Russian Academy of Sciences, Nizhny Novgorod, Russia, and a Lecturer with Nizhny Novgorod State University. His expertise include development of HIRETS and its application in medicine and hi-tech security systems. The current research focus of his department includes elaboration of near-field THz spectroscopy for analysis of biological systems. The Terahertz Spectroscopy Department is also involved with the development of THz devices for HIRETS, synthesizers of frequency detectors, highly stable generators based on QCL and superconductive Josephson generators. He has given more than ten invited talks on the related international conferences. His department is involved with several international projects concerning development and application of the THz technique and spectroscopy (e.g., NATO Science for Peace, TeraDec, and "Ultrafast Infrared Emitter on a Quantum Cascade" jointly with the Swiss National Foundation. He has authored and coauthored more than 240 papers.



M. Y. Li received the Ph.D. degree in radio physics from Nanjing University, Nanjing, China, in 2013.

After that, she continued her research with Chalmers University of Technology, Gotenburg, Sweden, as a Postdoctoral Researcher. Her research interests include terahertz receivers and emitters based on high-T_c materials.



Huabing Wang received the B.S. and Ph.D. degrees in radio physics from Nanjing University, Nanjing, China, in 1990 and 1995, respectively.

In 1995, he joined the Department of Electronic Science and Engineering, Nanjing University, Nanjing, China, as a Staff Member and was appointed an Associate Professor in 1996. In 1997, he became a Research Fellow with the Japan Science and Technology Corporation, working with the Research Institute of Electrical Communication, Tohoku University, Sendai, Japan, where he was promoted to an Associate Professor in 2001. After visiting the Universitaet Erlangen-Nuernberg, Germany, for one year, he spent two years as an ICYS Research Fellow with the National Institute for Materials Science, where he has been a Senior Researcher since 2006. He has been establishing laboratories for international research collaboration on terahertz solid-state devices at Nanjing University since 2011. His research interests are superconducting electronics (fabrication, characterization, design, and nonlinear dynamics of superconducting electronic devices) and terahertz applications.