Optimization of the CO_2 laser ran into difficulties because of the lack of understanding of the mechanism responsible for the production of inversion between vibrational levels in a gas discharge. The situation became clear with the emergence of a paper by N N Sobolev and V V Sokovikov [21] and Ref. [22] that followed. The authors compared data on the average electron energy in the discharge with the energy dependence of the excitation cross section for the corresponding molecular vibrational levels and saw that the electron impact mechanism is highly efficient. In this case, the inversion between vibrational levels is produced due to collisions with atoms and molecules. When these papers appeared, a start was made on a targeted theoretical description of the CO_2 laser and a new impetus was given to experiments aimed at its improvement. Notably, the USSR's first CO_2 gas-dynamic laser was put into operation in the framework of this work [23].

8. Conclusions

Summarizing the foregoing material, one may draw a conclusion on what underlay the success of Soviet science in the development of lasers.

• In the USSR there existed a large community of highly qualified scientists, which was permanently fed by scientific personnel from institutes of higher education (the Moscow Institute of Physics and Technology (MIPT), M V Lomonosov Moscow State University, the Moscow Engineering Physics Institute, etc.). From MIPT alone (MIPT was founded for preparing researchers in physics) several hundred graduates came to the LPI during the post-war years. The leaders of the laser program, N G Basov and A M Prokhorov, could rely on the scientific schools nourished by S I Vavilov, G S Landsberg, and L I Mandel'shtam at LPI.

• A system of financing the research had functioned in the USSR. The government responded to the needs of science and stimulated scientific progress. New facilities were commissioned to promote laser research: buildings on the LPI territory, branches of the LPI in Troitsk with accommodation for scientists, the Institute of Spectroscopy, the Polyus Scientific Production Association, and other laser research institutes.

• USSR industries were capable of providing the requisite components and instruments for research; in the USSR there were works for creating big research facilities and technologies for producing unique materials.

Notwithstanding the known shortcomings of the governance of that time, a strategically weighed program of scientific and technological development existed in the country. Laser research was a part of this program.

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Laser methods for generating megavolt terahertz pulses

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1. Introduction

The development of terahertz (THz) electromagnetic radiation sources is related to new methods and avenues of basic research in physics, chemistry, biology, and medicine, which have been making rapid strides during the last decade, as well as to new methods in different areas of applied research, including those related to new industrial technologies and security issues (see book Ref. [1] and references cited therein).

Terahertz radiation opens new paths and fresh unique possibilities for studying the properties and structure of substances and objects in the heretofore practically inaccessible spectral–temporal domain. Recording the probing terahertz pulses transmitted or reflected by an object and their subsequent amplitude–time and spectral analysis permits acquiring data about the object parameters and the substance properties in the terahertz range, as well as about the processes occurring therein, with a high (pico- and subpicosecond) temporal resolution. Along with use in basic research, terahertz pulses find practical applications in

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different applied areas. In recent years, in particular, several technical solutions have been proposed involving the use of terahertz radiation for microwave radar and the positioning of small-sized objects, recording microwave images of 'hidden' objects, analysis of pharmaceuticals, detection of explosives and drugs, and so forth [1].

According to the presently accepted classification, terahertz radiation (1 THz $= 10^{12}$ Hz) lies in the wavelength range from several millimeters to several dozen micrometers (from several hundred gigahertz to several dozen terahertz). Basic and applied research aimed at studying the properties of diverse objects and materials (including solids, liquids, gases, biological objects, etc.), investigating the methods of generation, propagation, and recording of terahertz radiation, and developing novel terahertz sources, terahertz vision, and radar systems is being successfully carried out in this rather broad and informative spectral range. These investigations are being pursued at leading universities and research centers in the USA, European Union countries, China, and Japan. In Russia, this work is being carried out, in particular, at M V Lomonosov Moscow State University, the Institute of Applied Physics of the RAS, the A M Prokhorov General Physics Institute of the RAS, the V A Kotel'nikov Institute of Radio Engineering and Electronics of the RAS, the Institute for the Physics of Microstructures of the RAS, the Institute of Spectroscopy of the RAS, several institutes of the Siberian Branch of the RAS, and St. Petersburg State University of Information Technologies, Mechanics, and Optics.

Terahertz research has been actively pursued since the late 1990s — early 2000s. This is due to the development of new, primarily laser-related, methods of generating pulsed and cw terahertz radiation, which enabled implementing efficient compact instruments and devices in practice.

The past years have seen the development and proposals of several methods of laser-driven generation of terahertz electromagnetic pulses (of the micro- and millimeter wavelength ranges). Among these methods are:

— the employment of ultrafast optoelectronic semiconductor switches of current and voltage (terahertz antennas, or the so-called Auston switches which were first proposed by D H Auston back in the 1970s [2]);

— the exploitation of the optical rectification effect (discovered by M Bass and P Franken [3] in 1962) and the generation of the difference frequency of ultrashort (pico- and femtosecond) laser pulses in nonlinear optical media;

— the harnessing of the spatial separation effect of charges of opposite signs (electrons and holes) occurring under optical excitation of semiconductors by pico- and femtosecond laser pulses, which is attended by the fast diffusive drift of current carriers in a thin surface layer of the material;

— the employment of quantum-cascade lasers and lasers that simultaneously generate two-wavelength radiation (carbon oxide lasers, and solid-state lasers with a broad amplification band).

Widely discussed in recent literature is the feasibility of obtaining high-intensity (with electric field amplitudes above 10^6 V cm^{-1}) ultrashort (pico- and subpicosecond) terahertz pulses and their application to the study of different nonlinear processes and phenomena in physics, chemistry, and biology, as well as in applied areas [1]. Among the rapidly advancing techniques of terahertz radiation generation in a broad frequency range (from several hundred gigahertz to several dozen terahertz) is the utilization of nonlinear interaction of femtosecond laser pulses with gaseous media (production of various laser-plasma objects, for instance, extended plasma channels — so-called filaments [1]), as well as of their interaction with nonlinear-optical crystals: the generation of terahertz laser pulses in $LiNbO_3$, ZnTe, GaAs, GaP, and GaSe crystals [1]. Another promising method of THz wave generation involves the development of new solid-state two-frequency lasers with the subsequent conversion of their difference frequency to terahertz radiation in nonlinear crystals or optoelectronic emitters.

Both of these techniques for generating pulsed electromagnetic radiation of ultrashort (picosecond) duration are being actively developed at the A M Prokhorov General Physics Institute of the RAS (RAS GPI).

2. Terahertz sources based on two-frequency lasers

Research aimed at developing new terahertz radiation sources, which is pursued at the RAS GPI, involves the production of two-frequency lasing in a diode-pumped solid-state laser with new high-efficiency active crystal media having broad overall amplification lines (gadolinium, yttrium, and mixed vanadates — Nd:$GdVO_4$, Nd:YVO_4, and Nd:$Gd_{0.7}Y_{0.3}VO_4$) and the subsequent conversion of the laser radiation to the terahertz spectral range by generating the difference frequency in the nonlinear GaP and GaSe crystals [4, 5].

Simultaneous two-frequency lasing is realized in the same active medium possessing a broad overall amplification contour (up to 5 nm in the 1-μm region), which is placed in a selective cavity of a diode-pumped solid-state laser. This approach does not necessitate additionally bringing two laser beams into coincidence and timing the operation of two independent lasers. We obtained two-frequency lasing in cw, Q-switching (nanosecond range), and mode-locking (picosecond range) regimes.

To maximize the peak output power of the two-frequency solid-state laser and, therefore, the peak power of the generated terahertz radiation, a picosecond Nd^{3+}:$GdVO_4$ laser with longitudinal diode pumping of the active medium was developed at the RAS GPI, which operated in a combined regime: with simultaneous Q-switching and active mode locking. This laser operating regime permits raising the peak output power by nearly two orders of magnitude in comparison with that for a picosecond laser operated only with active acoustooptical mode locking. The optical schematic of the laser is depicted in Fig. 1.

An Nd:$GdVO_4$ (0.5 at.%) laser crystal measuring $4 \times 4 \times 6$ mm was cut along the c-axis. The active element

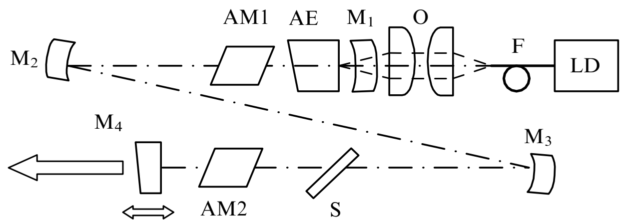

Figure 1. Schematic layout of a picosecond Nd^{3+}:$GdVO_4$ laser with simultaneous Q-switching and active mode locking: LD — laser diode pump; F — optical fiber (200 μm); O — objective; M_1 – M_4 — cavity mirrors; AE — active element; S — spectral selector (Fabry–Perot interferometer or Lyot filter); AM1 — acoustooptical Q-factor modulator, and AM2 — acoustooptical modulator intended for mode locking.

was pumped by an LIMO30-F200-DL808 linear laser diode array with a fiber radiation output ranging up to 25 W in power at a wavelength of 808 nm. The pump radiation was focused by an objective system which enabled obtaining a beam waist in the crystal from 150 to 400 μm in diameter. In experiments use was made of a Z-shaped cavity formed by four mirrors $M_1 - M_4$. The laser generated the principal TEM_{00} mode due to an iris placed into the cavity.

To actively Q-switch the laser, advantage was taken of an acoustooptical modulator (ML-321) controlled by a high-frequency sine-wave oscillator with a peak power of 30 W.

To achieve mode locking, use was made of an acoustooptical modulator (ML-202) with an output power of 8 W and a modulation frequency of 70 MHz, which corresponded to a laser pulse repetition rate of 140 MHz.

The duration of laser pulses was measured with the help of a streak camera with a resolution of 0.7 ps.

To obtain two-frequency output, spectral-selective elements (Fabry–Perot etalons) in the form of 120- and 83-μm thick plane-parallel YAG crystal plates were placed into the cavity. These selectors enabled obtaining the two-frequency radiation spaced respectively at 2.3 and 3.8 nm in wavelength, which in turn corresponded to the generated terahertz radiation with frequencies of 0.56 and 0.92 THz.

This laser system provided stable two-frequency generation of 80–120-ns long trains of picosecond pulses containing 15–20 separate 30–40-ps long pulses following one after another at a repetition rate of 140 MHz. The average output power for a train repetition rate of 10 kHz amounted to 350 mW. The laser system developed had stable radiation parameters which did not vary for several hours of continuous running, and was employed for high-efficiency nonlinear conversion of the laser radiation to the terahertz wavelength range in GaSe crystals. In this case, the output power of the terahertz radiation amounted to several microwatts, and the energy of the terahertz comb was as high as several tenths of a nanojoule. Of course, so low an energy level of the terahertz pulses does not by itself ensure attainment of the megavolt electric fields declared. That is why, the created two-frequency laser should be regarded as the master oscillator for a high-power laser system with an energy of more than several hundred millijoules, which is intended for the subsequent conversion of its output radiation to the terahertz range employing wide-aperture nonlinear GaSe crystals. Figure 2 displays a photograph of such a crystal 50 mm in diameter intended for operation together with the high-power two-frequency laser, which presently is under development at the RAS GPI.

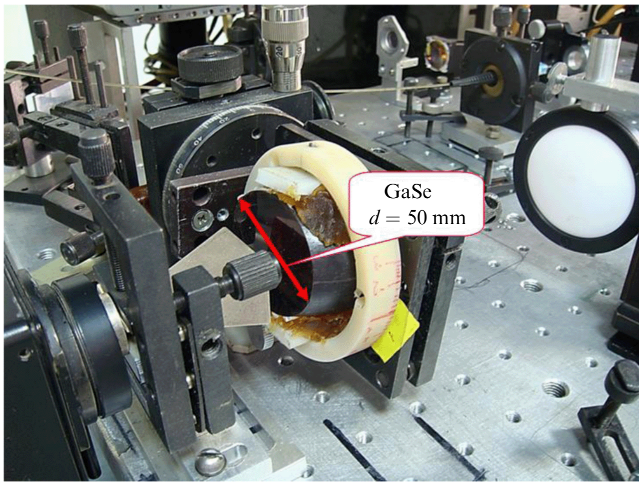

Figure 2. Terahertz converter module based on a GaAs crystal 50 mm in diameter, which is intended for operation together with the high-power two-frequency laser being developed at the RAS GPI.

3. Terahertz sources based on femtosecond lasers

Despite the long-standing successful employment of femtosecond lasers for the generation of terahertz radiation, the sources of terahertz pulses made on their bases (including commercially available ones) possess, as a rule, a low output power and a low peak intensity of the generated radiation. Typically, the generated terahertz pulses range from pico- to nanojoules in energy, their average power ranges between nano- and microwatts, and the intensities of the electromagnetic field lie in the range between several and several dozen kV cm^{-1}. Only with unique terahertz radiation sources relying on radically different generation techniques [free-electron lasers at the Budker Institute of Nuclear Physics, Siberian Branch of the RAS (Novosibirsk), the Stanford Picosecond Free Electron Laser Center (USA), the FOM-Institute for Plasma Physics (the Netherlands), etc.] has it been possible to obtain average power levels of up to several dozen watts and electromagnetic field strengths of several hundred kV cm^{-1}. Notably, the highest-energy (several dozen microjoules) and peak power (up to 100 MW) terahertz pulses were obtained in the generation of the transition radiation of relativistic electron beams of picosecond duration at the Brookhaven National Laboratory (USA). It is noteworthy that the uniqueness of such facilities (gigantic sizes and high operating costs) is a significant limitation for their wide application. As for the laser femtosecond sources under consideration, terahertz pulse energies of several dozen nanojoules and generated field intensities of several dozen kV cm^{-1} were until recently regarded as record high.

The low energy and peak power of femtosecond terahertz sources significantly limit their practical implementation. More specifically, terahertz radiation is largely attenuated in the propagation through the atmosphere due to the absorption by water vapor, which limits its practical use in radar systems for objects located at distances exceeding several dozen meters. Even in transparency windows (200–300 GHz), the attenuation is as strong as several dozen dB km^{-1}, with the consequence that the existing terahertz radiation detectors cannot ensure reliable signal detection, even at so short a distance.

However, the situation has changed radically in recent years due to the successful development and implementation of a new method of terahertz radiation generation, which is based on the optical rectification of femtosecond laser pulses with a pre-tilted wave front in stoichiometric MgO:$LiNbO_3$ crystals. This method was first proposed in 2002 by the Hungarian physicist J Hebling jointly with his colleagues from the Max-Planck-Institute for Solid State Researches in Stuttgart [6]. In 2007, in particular, scientists at the Massachusetts Institute of Technology (MIT) succeeded in reaching record high values of the energy parameters of terahertz pulses. Utilizing femtosecond laser pulses with an energy of 20 mJ, the MIT researchers obtained ultrashort terahertz pulses with an energy of up to 10 μJ and a peak power of 5 MW [7]. It was therefore demonstrated that the employment of femtosecond lasers makes it possible to generate pulsed

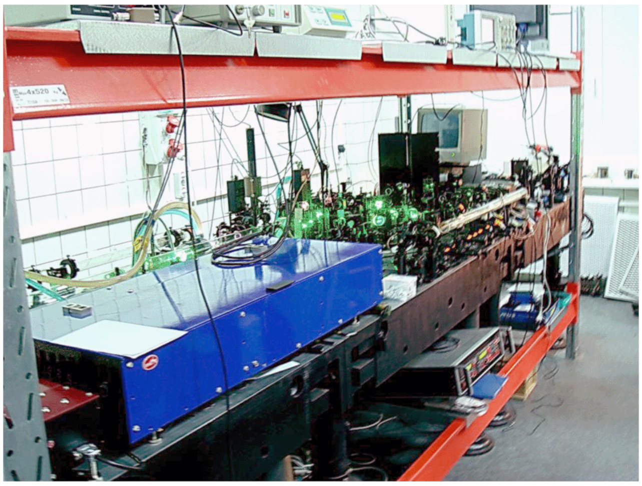

Figure 3. TERAFEM terawatt parametric laser complex.

terahertz radiation with a peak power which exceeds the peak power of the terahertz radiation generated by synchrotron sources and free-electron lasers. To state it in different terms, the technique of the optical rectification of femtosecond laser pulses with a transverse group delay in $MgO{:}LiNbO_3$ crystals enables obtaining the now record high energy efficiency of conversion of femtosecond laser pulses to terahertz radiation. (Specifically, the conversion efficiency of this technique is several orders of magnitude higher than the efficiency of conversion in ZnTe crystals, which are presently employed in the majority of terahertz radiation sources reliant on the optical rectification of femtosecond laser pulses.)

Femtosecond laser pulses with a tilted intensity front are required to meet the condition of their phase matching with the generated terahertz radiation. In lithium niobate crystals, the collinear matching of a wave interaction is not realized for the femtosecond laser radiation (e.g., of a titanium–sapphire laser) in use, and so Hebling et al. [6] proposed the application of noncollinear matching — the condition that the phase velocity V_{THz} of the generated terahertz wave is equal to the projection $V_{\text{las}} \cos\alpha$ of the group velocity vector of the laser pulse onto the former velocity direction, where α is the in-crystal angle between the propagation directions of the laser and the terahertz radiations. This condition may be fulfilled, in particular, for laser pulses with a tilted intensity front which is easily formed, for instance, when the laser radiation is obliquely incident on a diffraction grating [6]. Interestingly, this relation for noncollinear matching is similar to the relation for the propagation direction of the Cherenkov radiation generated by a dipole traveling at a supraluminal speed through a medium — the process which was first considered theoretically by V L Ginzburg [8] in 1959, and which was not realized experimentally until 45 years after the dawn of the laser era [9].

In 2009, a laser source of high-intensity terahertz pulses, which relies on the principle proposed in Ref. [6], was devised at the RAS GPI. The source possesses the highest energy parameters in Russia: a pulse power of over 1 MW, and a field intensity above 1 MV cm^{-1}. The terahertz source comprises a terawatt laser complex (TERAFEM), whose overall view is shown in Fig. 3, and a module for generating megavolt terahertz electromagnetic pulses, whose optical scheme is depicted in Fig. 4. The peak power of the laser complex based on parametric radiation amplification [10] amounts to 1 TW at the central wavelength (910 nm) for a pulse duration of 45 fs. The module intended for the generation of megavolt terahertz pulses relies on the optical rectification of femtosecond laser pulses with a tilted intensity front in wide-aperture (measuring $30 \times 10 \times 10$ mm) magnesium-doped stoichiometric lithium niobate ($MgO{:}LiNbO_3$) crystals. For a laser pulse energy of 30–40 mJ, the energy of the terahertz pulses produced by the module amounts to 2–3 μJ; after focusing them on a spot 500 μm in diameter, the resultant electric field amplitude of the terahertz wave field exceeds 10^6 V cm^{-1}.

The high-intensity terahertz radiation system under consideration is intended for studying extreme states of matter in the terahertz spectral domain and for solving a number of applied problems. In this case, the laser part of the complex is independently employed for executing experiments in the production of charged particles and the generation of X-rays, and in studies of the mechanisms of plasma formation and the filamentation of laser radiation.

4. Techniques for characterizing high-intensity terahertz pulses

In the work carried out to develop pulsed terahertz sources, special emphasis was placed on the techniques and means for measuring their energy and time parameters.

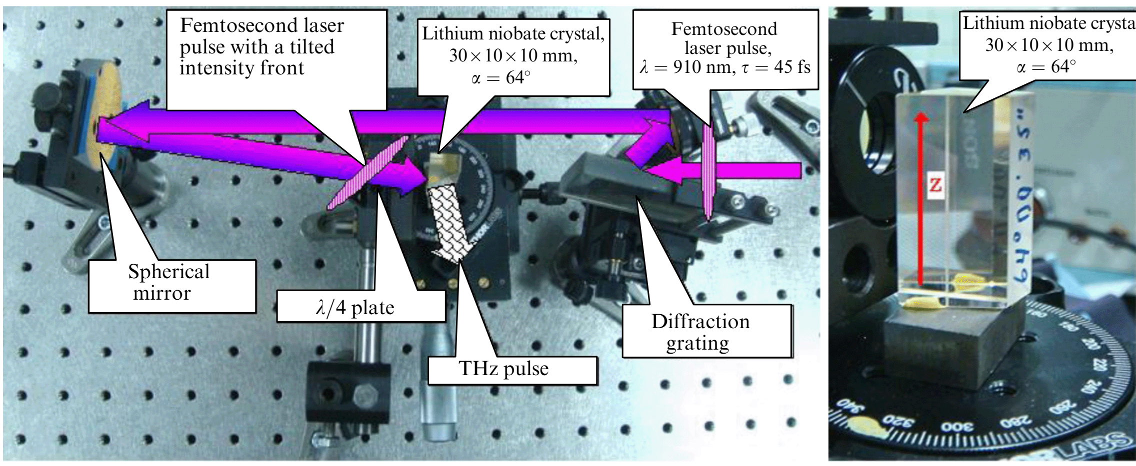

Figure 4. Optical scheme of the module for generating megavolt terahertz electromagnetic pulses. External appearance of a stoichiometric lithium niobate crystal (Z is the direction of the crystal axis).

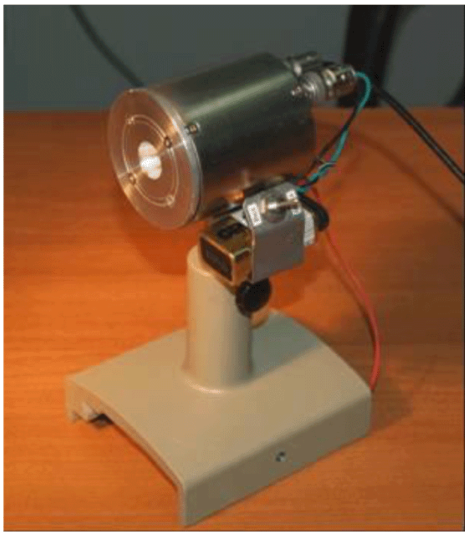

Figure 5. Pyroelectric terahertz pulse energy meter.

Table 1. Parameters of pyroelectric terahertz pulse energy meters.

Parameter	Value
Spectral range	0.1 – 3 THz
Responsivity to single THz pulses	$S_{\mathrm{pulse}} = 1.9 \times 10^6$ V J^{-1} (±15%)
Responsivity for 22-Hz modulation frequency and 1-MΩ load	20,000 (±15%) V W^{-1} at 0.14-THz frequency 16,000 (±15%) V W^{-1} for broadband radiation in 0.2–2 THz frequency range
Dynamic range	0.1 μW – 0.35 mW
Noise level	1.0 mV

To record the energy parameters of terahertz pulses, high-responsivity (1.9×10^6 V J^{-1}) broadband (spectral range: 0.1–3 THz) pyroelectric sensors with a linear dynamic range of more than 10^3 and a detection threshold of 1 nJ were developed, fabricated, and calibrated. An external appearance of the pyroelectric terahertz pulse energy meter is given in Fig. 5, and its parameters are collected in Table 1.

To measure the amplitude–time profile of ultrashort (~ 1 ps) single (repetition rate close to 1 Hz) terahertz pulses, the method of spatial visualization of the terahertz field was selected and implemented, which is based on the electrooptical recording of the 'image' of a *single* terahertz pulse with a gating *single* femtosecond laser pulse due to the optical anisotropy in an electrooptical ZnTe crystal, induced by a terahertz field.

Figure 6 shows the schematic diagram of the method and the results of measurement of the temporal profile of a picosecond terahertz pulse.

5. Conclusions

Laser sources of pulsed terahertz radiation of ultrashort duration were devised in the laboratories of the A M Prokhorov GPI of the RAS, which make it possible to obtain record high electric field intensity — over 10^6 V cm^{-1}. The achieved megavolt intensity level of the terahertz field opens up exciting possibilities for a new line of investigation in physics — the nonlinear optics of terahertz waves, which has recently begun to progress rapidly due to the advent of new

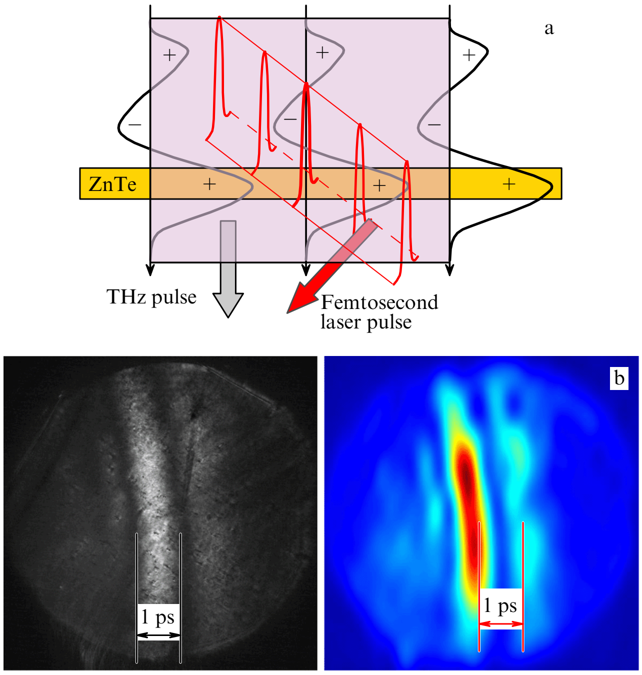

Figure 6. (a) A schematic of the technique of spatial visualization of a terahertz field. (b) Recorded temporal profiles of single terahertz pulses.

compact laser sources of high-intensity terahertz pulses. Among the problems solvable by the methods of nonlinear terahertz optics, mention should be made, in particular, of nonlinear plasma production mechanisms — ionization of substances and the 'optical' breakdown of material media; the generation mechanisms of higher-order harmonics of currents and radiation; charged particle acceleration by high-intensity terahertz fields; modulation and parametric plasma instabilities in the field of terahertz radiation, and self-focusing of terahertz pulses.

Fundamentally, the technique of the optical rectification of femtosecond laser pulses with a tilted intensity front in nonlinear crystals permits generating single-cycle picosecond terahertz pulses with an even higher energy (over 100 μJ) and higher field intensity amplitude (up to 10^9 V cm^{-1}). Such pulses may be obtained in the presently existing multiterawatt laser facilities, in particular, in the Luch facility at the Institute of Laser Physics Research of the Russian Federation Nuclear Center 'All-Russian Research Institute of Experimental Physics' (Sarov).

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The early history of the injection laser

Yu M Popov

One of the spectacular achievements of world science, which strongly affected the technological level of modern society, is the generation of optical radiation in semiconducting materials. The concept of a semiconductor laser and the first studies in this area were initiated at the Lebedev Physical Institute, RAS (FIAN in *Russ. abbr.*), where a group of young scientists organized by N G Basov was engaged, beginning from 1957, in the problem of creating a semiconductor laser. These were pioneering investigations not only in our country but also in the world, along with the studies of C Townes and A Schawlow in the USA.

Although molecular generators operated on gases, the paramagnetic amplifiers of stimulated emission already utilized crystals, confirming the possibility of obtaining inverted population in solids. Semiconductors as active media attracted our attention because they have high absorption (amplification) coefficients. This opened up the possibility of constructing resonators of a small size. At FIAN, the properties of semiconductors in strong electric fields were studied at the Laboratory of Semiconductor Physics headed by B M Vul. N G Basov and his collaborators discussed with B M Vul and his colleagues the possibility of obtaining the inverted population required for light amplification in semiconductors. In 1958, N G Basov, B M Vul, and Yu M Popov filed an application for a patent and published a paper on using short current pulses for the avalanche multiplication of charge carriers from the valence band (or from impurities), and producing the inverted population due to cooling carriers by a lattice after the instant removal of the field [1]. The patent was recorded in the State Register by the Committee on Inventions and Discoveries of the Council of Ministers of the USSR on 7 July 1958. N G Basov reported this work at the first conference on quantum electronics in the USA in 1959.

Because the production of interband avalanche multiplication in germanium and silicon required high electric field strengths, whereas optical interband transitions were indirect, we decided to experimentally study the ionization of impurities in these materials and to use narrow-gap semiconductors with direct optical transitions for interband ionization. At that time, the best studied semiconductor of this type was indium antimonide which was grown at the Leningrad Physicotechnical Institute (LFTI in *Russ. abbr.*) at the laboratory headed by D N Nasledov, with whom we made friends. A group at the FIAN Laboratory of Semiconductor Physics studied the recombination emission of electrons ionized from shallow impurities in germanium, while a group at the Laboratory of Oscillations investigated emission observed during avalanche multiplication in indium antimonide. However, although some interesting results were obtained in these studies, no evidence of stimulated emission was observed. The main difficulties were the requirement to obtain current pulses with very short fronts and the complexity of performing measurements in the infrared region. Later on, already after the creation of semiconductor lasers by other methods, the inverted population between the energy levels of donor impurities in silicon was achieved using emission from a CO_2 laser.

In 1960, we published a long paper in *Usp. Fiz. Nauk* (*Sov. Physics–Uspekhi*) presenting both a review of the main methods and media for the creation of lasers and a number of original concepts about the use of semiconductors for this purpose [2]. In particular, the inverted population was formulated as a condition for the nonequilibrium distribution functions in bands, and we proposed forming a resonator by means of parallel output facets of semiconducting crystals having reflection high enough for providing the feedback. At the beginning of 1961, we proposed a method for producing the inverted population in semiconductors by fast electrons, and in March the concept was suggested and the main conditions were formulated for bringing about the inverted population by injecting nonequilibrium charge carriers through the p–n junction in degenerate semiconductors [3]. As a result, the fundamentals of the theory of semiconductor lasers were developed by the early 1960s. Consider in detail paper [3] where the possibility of creating an injection (diode) laser was proposed and substantiated in the world first (the paper was submitted to *JETP* on 18 April 1961 and published in June 1961). The main results of paper [3] are as follows.

(1) When a forward voltage is applied to the p–n junction in a semiconductor, the concentration of minority charge carriers near the p–n junction increases due to a decrease in the potential barrier formed by a spatial charge in the p–n junction. The maximum concentration of these carriers corresponds to the complete removal of the potential barrier by the external electric field and is on the order of their concentration in the part of the crystal where they are majority charge carriers (we assume that the p–n junction is sharp). The negative temperature in interband transitions appears only when the Fermi quasilevels corresponding to the nonequilibrium electron and hole concentrations satisfy the following condition

$$F_{\mathrm{e}} + F_{\mathrm{p}} > \Delta\,, \tag{1}$$

where F_{e} and F_{p} are the Fermi quasilevels for electrons and holes, and Δ is the band gap. When a forward voltage is applied to the p–n junction, the Fermi quasilevel of minority carriers near the p–n junction will be close to the Fermi level in the part of the crystal where they are majority. In this case, it follows from inequality (1) that the carriers should be degenerate at least in one part of the p–n junction. Semiconductors with such p–n junctions are termed tunnel

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