VILNIUS UNIVERSITY
CENTER FOR PHYSICAL SCIENCES AND TECHNOLOGY
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ANDRIUS BIČIŪNAS

SEMICONDUCTOR MATERIALS FOR COMPONENTS OF OPTOELECTRONIC TERAHERTZ SYSTEMS ACTIVATED BY FEMTOSECOND 1 μm WAVELENGTH LASER PULSES

Summary of doctoral dissertation

Physical Sciences, Physics (02P), Semiconductor Physics (P 265)

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PUSLAIDININKINIŲ MEDŽIAGŲ, SKIRTŲ 1 μm BANGOS ILGIO FEMTOSEKUNDINIAIS LAZERIO IMPULSAIS AKTYVUOJAMŲ TERAHERCINIŲ OPTOELEKTRONIKOS SISTEMŲ KOMPONENTAMS, TYRIMAS

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1. Introduction

Terahertz (THz) pulse generation and detection using optoelectronic semiconductor components excited by femtosecond laser pulses has become lately a powerful experimental technique finding numerous applications in THz time-domain spectroscopy and THz imaging [1]. Traditionally, mode-locked Ti:sapphire lasers emitting at the wavelengths around 800 nm are used for this purpose because they are more advanced technically and commercially more mature than other similar products and because their photon energy perfectly matches the absorption band of GaAs. Nonstoichiometric GaAs obtained either by molecular-beam-epitaxy growth at reduced temperatures (low-temperature-grown (LTG) GaAs) [2] or by high-energy, heavy ion implantation [3] is characterized by unique set of material parameters [4] that facilitates its applications in pulsed THz components. However, because Ti:sapphire lasers require a rather complicated, many-stage optical pumping arrangement, these systems are quite bulky and complicated, which prevents their wider applications.

One solution of this problem would be using the lasers emitting in the spectral ranges close to 1 or 1.55 μm, which can be directly pumped by diode laser bars. Recently, several compact, efficient and cost-effective solid-state and fiber laser systems that generate femtosecond pulses at these near-infrared wavelengths have been developed [5, 6] and have been employed for activating THz time-domain spectroscopy systems [7]. The main obstacle in further development of these systems is the absence of material with appropriate bandgap and parameters high dark resistivities and subpicosecond carrier lifetimes – similar to those of LTG GaAs, which could be used for manufacturing photoconductive THz emitters and detectors. In the case of THz detectors activated by 1 μm wavelength femtosecond lasers, epitaxial layers of LTG InGaAs [7] or LTG GaBiAs [8] were employed, whereas no material appropriate for photoconductive THz emitters has been identified as yet.

This work is focused on the THz pulses generation in semiconductors surfaces and on using photoconductive THz emitter activated by femtosecond 1 μm wavelength laser pulses.

Main goal

1. The main goal of this dissertation was to develop and to investigate semiconductor materials for terahertz (THz) pulse emitters to be used in Terahertz time-domain spectroscopy (THz-TDS) systems using a 1 μm wavelength femtosecond laser radiation.

The objectives of the study

1. To compare the efficiency of various semiconductor surface THz emitters photoexcited by femtosecond 1 μm wavelength laser pulses.
2. To explore the physical mechanisms of terahertz generation from narrow gap semiconductors (InAs, Ge, InSb) surfaces.
3. To produce and to investigate ultrafast photoconductors from low-temperature grown (LTG) GaBiAs layers and to adapt them for THz pulse generation.
4. To explore the THz pulse generation and detection using a 1 μm femtosecond laser radiation and photoconductors manufactured from LTG GaAs.

The novelty of the study

- THz pulse emission from the surfaces of various semiconductors illuminated by 1 μm wavelength femtosecond laser pulses have been studied systematically.
- Azimuthal angle dependences of terahertz pulse emission from low symmetry, (112)-cut InSb samples were measured for the first time. It has been shown that both nonlinear optical effects (optical rectification and electric field induced optical rectification) are necessary to take into account when explaining the THz signal generation from the surfaces of this material.
- New LTG GaBiAs, LTG GaAs emitters and LTG GaBiAs detectors for THz-TDS systems based on femtosecond 1 μm wavelength laser have been developed and THz pulse emission from GaBiAs and LTG GaAs emitters have been thoroughly investigated.

- Using the THz-TDS system based on photoconductive antennae manufactured from GaBiAs layers and activated by low average power, 1030 nm femtosecond Yd-doped fiber laser record high optical-to-THz power conversion efficiency $\sim 5\times10^{-4}$ was obtained.

Points to be maintained (statements of defense)

1. The electric field in inverse layer at the p-type InAs crystal surface induces optical rectification of femtosecond laser pulses, thus p-type InAs crystals are efficient THz pulse emitters.
2. It is possible to estimate quantitatively the influence of optical rectification and electric field induced optical rectification by investigating the surface generated THz pulse amplitude dependence on azimuthal angle measured on (112) or other low symmetry planes.
3. The photoconductors made from epitaxial GaBiAs layer allow an efficient conversion 1 μm wavelength femtosecond pulses into terahertz radiation due to its unique combination of high electron mobility (~2000 cm^2/Vs) and picosecond lifetimes.

1. Terahertz emission from semiconductor surfaces illuminated by femtosecond Yb:KGW laser pulses (P1, P2, P3)

It is known that THz radiation can be emitted from unbiased semiconductor surfaces photoexcited by femtosecond laser pulses [9]. At Ti:sapphire laser wavelength this effect is most efficient when using p-type InAs crystals [10]. Here we present a systematic study of THz pulse emission from the surfaces of various semiconductors illuminated by femtosecond Yb:KGW (potassium gadolinium vanadate) laser.

1.1 Experimental details

As the optical pulse source for THz pulse generation and sampling the oscillator from a high repetition rate femtosecond laser system Pharos (Light Conversion Ltd) was used. The oscillator based on directly diode-pumped Yb:KGW (KGa(WO3)) crystal. Kerr-lens mode-locking was used to generate optical pulses with the central wavelength of 1030 nm. The average output power of the oscillator was 2 W, the pulse duration was 70 fs (Gaussian fit), the spectral width (FWHM) was ~22 nm, and the pulse repetition rate was 76 MHz. Different semiconductor surfaces have been illuminated by the laser beam at the angle of 45° and THz radiation emitted from these surfaces was monitored in quasi- reflection direction. Azimuthal angle dependences of the emitted THz pulse amplitude were measured for all investigated semiconductors by rotating the crystal around the normal to its surface. All measurements were performed at room temperature.

For THz pulse detection the LTG GaBiAs detector was used. Epitaxial GaBiAs layer was grown on semi-insulating (100) oriented GaAs substrate in a solid-state molecular-beam-epitaxy (MBE) system at 280°C substrate temperature at the growth rate of 2 μm/h. The thickness of the layer was 0.4 μm; it has been used for manufacturing coplanar Hertzian dipole antennas with 15 μm wide photoconducting gap between Ti–Au electrodes and equipped with a hemispherical lens made from a high resistivity Si. S- and P-polarized THz signals were separated by rotating the detector that was polarization sensitive and by inserting at a Brewster angle a semi-insulating InP wafer into the THz beam path. The performance of the experimental system is illustrated in Fig. 1, where

THz pulse and its corresponding Fourier spectrum radiated from the surface of p-type InAs crystal are shown.

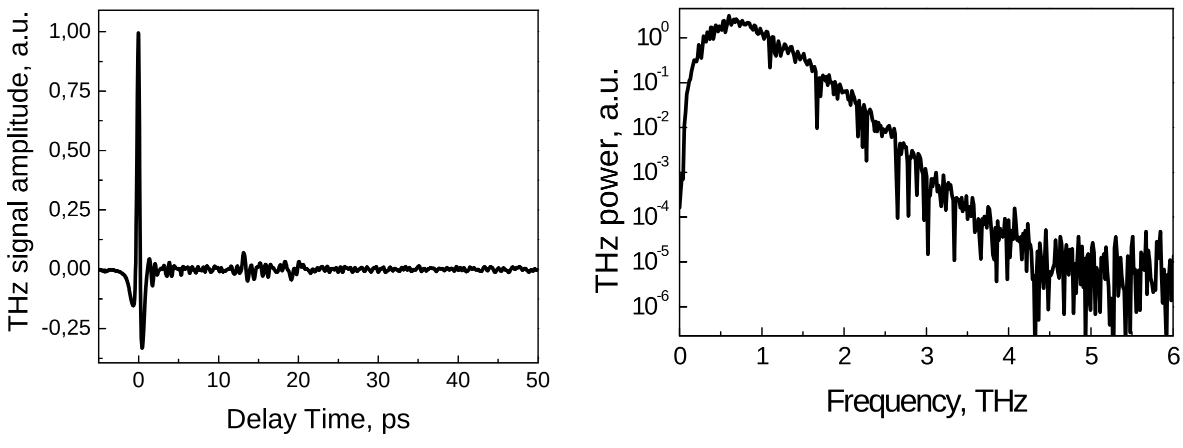


Fig. 1. THz pulse (a) and its Fourier spectrum (b) radiated from the surface of p-type InAs crystal and measured using GaBiAs detector.

Besides of p- and n-type InAs crystals, THz emission from the surfaces of InSb (intrinsic conduction at the room temperature) and p-and n-type Ge were investigated. InAs and Ge crystals were cut parallel to the (111) crystallographic plane. The InSb crystals were cut parallel to the (111) and (112) crystallographic planes. Moreover, epitaxial layers of $Cd_xHg_{1-x}Te$ (with x = 0.2 and 0.3) grown on CdTe substrates, and $In_xGa_{1-x}As$ (with x = 0.2 and 0.5) on InP substrate with the symmetry of (100) were investigated. The same symmetry had the wafers of semi-insulating GaAs that were also investigated in the experiments.

1.2 Results and discussion

Diagram presented in Fig. 2 compares THz pulse amplitudes as emitted from the surfaces of various semiconductor materials under the same experimental conditions. Most efficiently THz pulses are generated at the surface of p-InAs, followed by several other narrow-gap semiconductors. There are two main groups of the physical mechanisms leading to THz emission from semiconductor surfaces excited by femtosecond laser pulses: surface photocurrent surge and optical rectification of femtosecond laser pulses due to their nonlinear optical interaction with the material [11].

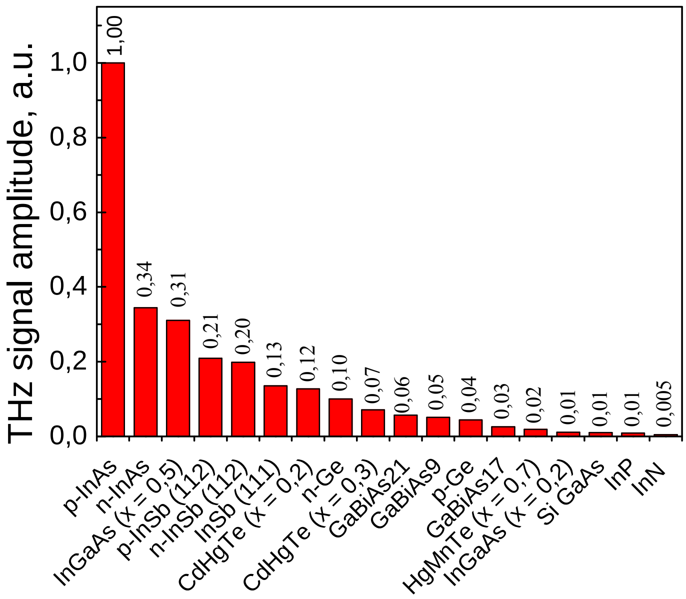


Fig. 2. Relative comparison of THz pulse amplitudes, emitted from the surface of various semiconductors illuminated by femtosecond 1.03 μm wavelength laser pulses. Light intensity on surface emitters is 0.63 W/cm^2.

Standard way to separate these mechanisms is measuring azimuthal angle dependences of the radiated THz pulse amplitudes: for semiconductor crystals that, typically, have a cubic symmetry the photocurrent is isotropic, whereas THz pulses radiated due to the optical rectification effect exhibit complex dependences on the azimuthal angle determined by the symmetry of the photoexcited crystallographic plane and on the order of the optical nonlinearity (second or third order) that causes this effect [12]. In the following we will discuss the azimuthal angle dependences of THz radiation from different semiconductors separately.

1.2.1 InAs crystals

When excited by Ti:sapphire laser pulses, InAs narrow-gap semiconductors are the best THz pulse emitter because of large difference in the velocities of photoexcited electrons and holes that causes efficient spatial separation of these current carriers and because of a significant third order optical nonlinearity leading to the EFIOR (Electrical Field Induced Optical Rectification) effect at the surface of these materials [11]. Similar features of this effect were observed also when femtosecond 1030 nm wavelength optical pulses were used for the photoexcitation.

Fig. 3 shows azimuthal angle φ dependences of P- and S-polarized THz pulse amplitude measured on two differently doped InAs crystals.

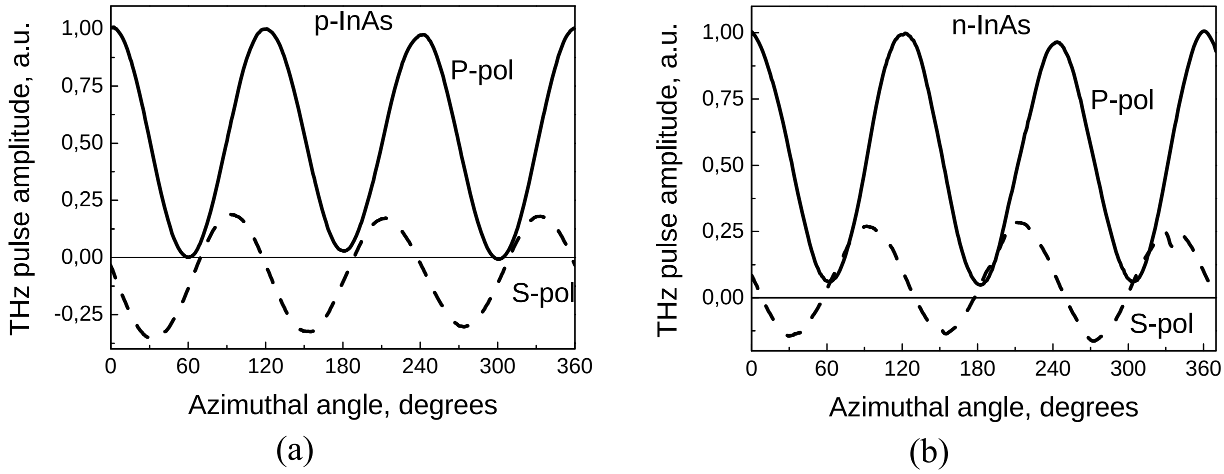


Fig. 3. Azimuthal angle of P- (full lines) and S-polarized (dashed lines) THz pulse amplitudes emitted from the (111) surfaces of p-InAs (a) and n-InAs (b) crystals.

All curves show the variation proportional to cos(3φ) that is typical for the optical rectification effect on (111) surfaces; the presence of S-polarized THz radiation, which cannot be generated by any mechanisms related to the photocurrent surge, is also evidencing the significance of the nonlinear optical effects. As in the case of 850 nm excitation [12], the most probable cause of THz emission is the optical rectification induced by the electric field that is appearing at the crystal surface due to the photoexcited electron and hole separation. THz pulses radiated from p-type InAs surface have more than twice larger amplitudes than those radiated from n-InAs. This difference can be explained by the presence of a built-in electric field of the inversion layer at p-InAs surface [10]. Surface electric field in p-InAs is of the same polarity as the field arising due to the photoexcited electron and hole separation; however, it interacts with the whole optical pulse and not with its part as in n-InAs, where optical rectification effect is due to the electric field induced by the movement of the electrons excited during the initial phase of the laser pulse.

This conclusion about the mechanisms of THz emission from n- and p-type InAs is supported by the measurements of THz pulse (S-polarized) amplitude dependences on the laser beam intensity presented in Fig. 4.

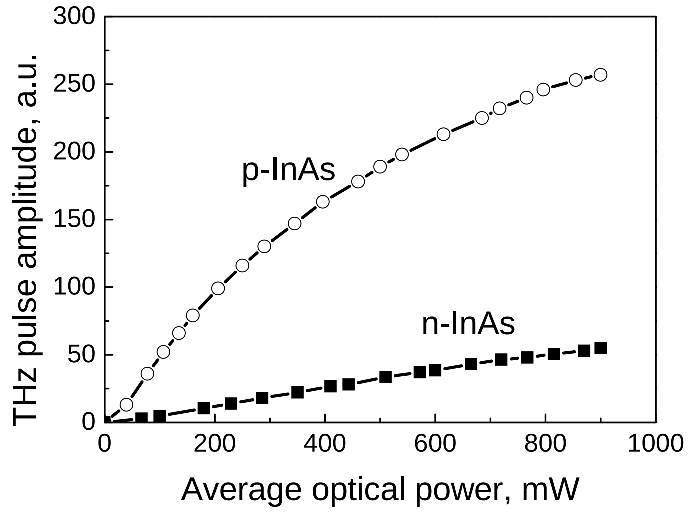


Fig. 4. THz pulse amplitude as a function of the average optical power for two InAs crystals.

As can be seen from fig. 4, the dependence measured on p-InAs shows a strong tendency to saturation, whereas in the case of n-InAs it remains linear to the largest intensities employed. This difference can arise due to the built-in surface field screening in the former crystal by carriers generated during the slowly varying initial part of the laser pulse.

1.2.2 GaAs and InGaAs samples

The symmetry of the surfaces of semi-insulating GaAs crystal and epitaxial $In_{0.5}Ga_{0.5}As$ layers corresponded to (100) crystallographic planes for which the EFIOR effect has only an azimuthal angle independent component. Therefore, azimuthal angle dependences of THz pulse amplitude proportional to cos(2φ) that were experimentally observed on the samples of these materials (Fig. 5) evidence the presence of the second-order nonlinear optical rectification (OR) effect [13].

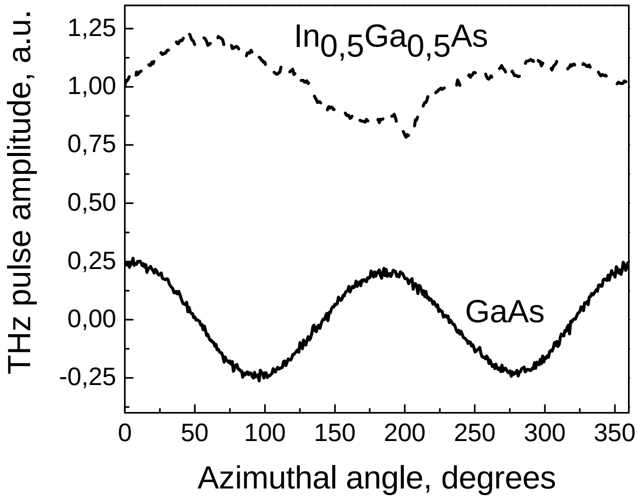


Fig. 5. Azimuthal angle dependences of P-polarization THz pulse emitted from (100) planes of GaAs (full line) and $In_{0.5}Ga_{0.5}As$ (dashed line).

For the case of GaAs transparent to 1030 nm wavelength radiation, OR effect is the only physical mechanism leading to THz emission. For $In_{0.5}Ga_{0.5}As$ that is absorbing laser radiation, besides the optical rectification contribution there is a significant azimuthal angle independent component, which can be caused by the EFIOR or/and photo-Dember effects.

1.2.3 InSb crystals

Azimuthal angle dependences of THz signal radiated from the surfaces of InSb (Fig. 6) are similar to those measured on InAs (Fig. 3) but the amplitudes of THz pulses are smaller. Although the excess energy of photoexcited electrons in InSb is larger than in InAs, which has a larger energy bandgap, electric field induced by the electron and hole separation is weaker because of the intense electron scattering to subsidiary conduction band valleys laying at L and X points of the Brillouin zone [14].

Fig. 6 show that both the photo-Dember and the nonlinear OR contribute to terahertz generation. Is known that for cubic crystals and (111) crystallographic plane, both OR and EFIOR effect contributions are changing proportionally to cos(3φ), whereas for the (100) plane, the EFIOR effect contribution does not depend on φ and the OR effect contribution varies proportionally to cos(2φ). Because it has been found experimentally that terahertz pulse amplitudes radiated from both those planes of InSb

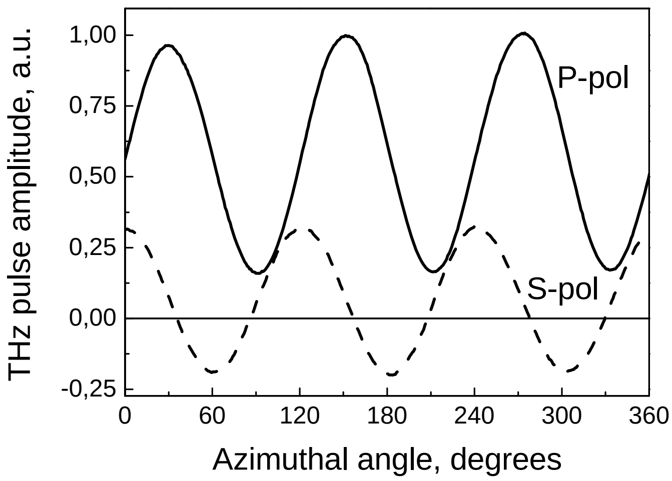
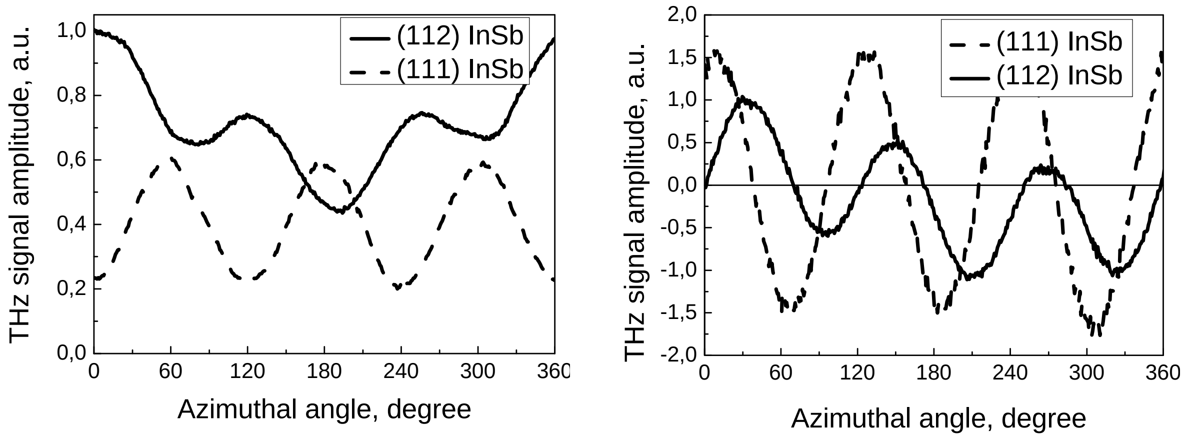


Fig. 6. Azimuthal angle of P- (full line) and S-polarized (dashed line) THz pulse amplitudes emitted from the (111) surface of InSb.

crystal depend on the azimuthal angle [14] it evidences unambiguously that at least the OR effect is contributing to the terahertz emission from this material. The role of the EFIOR effect, which is playing a very important role in InAs, cannot be identified from the measurements performed with the samples of two crystallographic orientations indicated above.

In this work, we have investigated the azimuthal angle dependences of terahertz pulse emission from lower symmetry, (112) crystallographic plane InSb samples. Fig. 7 compares the azimuthal angle dependences of *P*- and *S*-polarized terahertz pulse amplitudes measured on the (112) and (111) crystallographic planes of InSb. The curves are normalized to their maxima for (112)-cut crystals. For (112) crystallographic plane, the maximum amplitude of *P*-polarized terahertz pulse is approximately ten times larger than the largest magnitude of *S*-polarized terahertz radiation pulse. The angular dependences of terahertz radiation from (111) planes show the expected proportionality to sin(3φ), whereas the dependences measured on (112) plane have a more complicated structure. The curve corresponding to a *P*-polarized terahertz signal has one clear maximum at the (112) crystallographic direction and one minimum for the opposite direction; the angular dependence of *S*- polarized terahertz signal, on the other side,



(a) (b)

Fig. 7. Azimuthal angle dependences of *P*- (a) and *S*-polarized (b) terahertz pulse amplitudes measured on (112) (solid line) and (111) (dashed line) crystallographic planes of InSb. The curves are normalized to their maxima for (112)-cut crystals.

beside the sin(3φ) component has a clearly distinguishable component proportional to sin(2φ). It is important to point out that the largest *P*-polarized terahertz signals generated from the (112) plane have ~1.5 times higher amplitude than those generated from the (111) plane. In the case of *S*-polarized terahertz signals, this relation is opposite.

Theoretically it has been shown that for (112) crystallographic plane the contributions of both OR and EFIOR effects depend on the azimuthal angle and these dependences are different. Experimental terahertz pulse amplitude dependences on the azimuthal angle were compared with the theory; it has been shown that both nonlinear optical effects are necessary to take into account when explaining the experimental observations ($EFIOR/OR \approx -1.92.$).

1.3 Conclusions

In conclusion, we have compared the amplitudes of THz pulses emitted from the surfaces of various semiconductors after their excitation by femtosecond 1030 nm laser pulses. It has been found that this effect is most efficient in p-type InAs. THz time-domain spectroscopy system utilizing p-InAs emitter and detector made from low-temperature grown GaBiAs, when activated by 70 fs duration laser pulses, has demonstrated the signal-to-noise ratio of 60 dB and the bandwidth of 5 THz. In the majority of the investigated semiconductors the main contribution to THz pulse emission comes from the optical rectification effect induced by the simultaneous action of the third-order optical nonlinearity and the electric fields arising due to photoexcited electron and hole separation and/or the band bending at the semiconductor surface.

Also we have investigated the azimuthal angle dependences of terahertz radiation emitted from photoexcited (112) surfaces of InSb crystals. These dependences had a more complicated shape than the curves measured on crystalline planes of a lower symmetry allowing the separation of two nonlinear optical mechanisms of terahertz emission: OR and EFIOR. It has been found from the comparison of the experimental measurements with the theory that both these effects contribute to the terahertz emission with opposite signs of terahertz signal, and the EFIOR effect contribution is approximately two times larger. It has also been shown that maximum terahertz pulse amplitudes generated from (112) planes are 1.5 times larger than those emitted from (111) planes. It can be assumed that further enhancement of terahertz signals is possible if crystallographic planes with even lower symmetry would be photoexcited by femtosecond laser pulses.

2. Terahertz time-domain spectroscopy system based on femtosecond 1 µm wavelength laser and GaBiAs photoconducting components (P4, P5)

$GaBi_xAs_{1-x}$ epitaxial layers with a few percent of Bi atoms grown on GaAs substrates have, recently, become a subject of an enhanced interest because of their unusual physical properties [15], including a large bandgap reduction (~60 meV per percent of Bi) and a strong enhancement of the spin-orbit splitting after the incorporation of Bi [16]. These properties make the diluted bismides attractive candidates for GaAs-based applications in long-wavelength optoelectronic as well as in spintronic devices. In the case of $GaBi_xAs_{1-x}$ the conduction band transport is expected to be much less perturbed than in GaN_xAs_{1-x}, a material with analogously large bandgap reduction, for which the degradation of the electron mobility has turned to be detrimental for applications in optoelectronic devices such as multijunction solar cells.

2.1 Experimental details

Epitaxial $GaBi_xAs_{1-x}$ layers were grown on semi-insulating (SI) GaAs (100) substrates in a solid-state MBE system. The substrates were bonded to a molybdenum holder using an indium solder; their temperature was measured by a thermocouple fixed on the back side of the holder. $GaBi_xAs_{1-x}$ layers were grown at temperatures of 250-280 °C and As_4/Ga beam equivalent pressure ratio of ~3. No post growth annealing was used.

Four $GaBi_xAs_{1-x}$ samples were investigated with the energy bandgaps determined from the spectral absorption measurement. GaBiAs layers bandgaps were from 0,95 eV (sample GaBiAs26 and GaBiAs35) to 1.1 eV (sample GaBiAs16) corresponding to the BiAs part in the alloy of from $x \approx 0{,}04$ to $x \approx 0{,}08$. Hall effect measurements was performed at room temperature have shown that the resistivity of the layers was exceeding 2000 Ωcm, i.e. much larger that it has been achieved when growing at higher temperatures [17]. In contrast to the results of [17], where p-type conduction of GaBiAs was evidenced, the sign of the Hall-voltage was negative. Strong temperature dependence of the resistivity as well as non-realistically low electron mobility as determined from the Hall measurement has led us to the assumption that the samples are highly compensated and are of a nearly intrinsic conduction.

Photoconductivity dynamics in GaBiAs samples was investigated by using optical pump-THz probe technique. Experimental set-up was based on Yb:KGW laser (the pulse duration of 70 fs, the central wavelength of 1030 nm, and the pulse repetition rate of 76 MHz) and photoconductive THz emitter and detector pair manufactured from LTG GaBiAs. A 1.9 mm diameter pinhole was used to overlap THz and optical pump beams on the sample. To maximize the transmitted THz power through the pinhole, the THz beam was focused before and collimated after the pinhole by two hemispherical Teflon lenses. For investigating the conductivity dynamics in the samples, the amplitude of THz transient at its maximum was measured as a function of the optical delay between the pump pulse and the parts of the laser pulses exciting THz emitter and THz detector.

Another set of experiments was performed with a home-built positive dispersion Yb–doped fiber oscillator based on the dual-output scheme introduced by Chong *et al.* [18]. The mode locking in this oscillator is based on nonlinear polarization evolution; an interference filter is included for spectral amplitude and phase shaping; and the laser operates in an all-positive dispersion regime. The repetition rate of the oscillator is 45 MHz. Beams with average powers of ~8 mW and ~11 mW were used for the photoexcitation of the THz emitter and the detector, respectively. Spectral tenability of the oscillator output in the range of 1030–1050 nm was achieved by tilting the interference filter in the free space gap of the fiber loop. The output pulses were compressed to the duration of ~160 fs in a double-pass compressor based on two 900 lines/mm transmission gratings. For the measurement of the dependence on the pulse duration, the latter was tuned by changing the distance between the diffraction gratings in the pulse compressor. The temporal profile and phase of the generated pulses was characterized with a home-built second harmonic frequency resolved optical gating device.

The measurements were performed with optimally compressed laser pulses, and with chirped pulses of both positive and negative chirp. Because of the interplay of the higher orders of spectral phase, negatively chirped pulses acquire a double-peak temporal structure, whereas positively chirped pulses retained a single-peak temporal shape. In the following, only results corresponding to the positively chirped and fully compressed optical pulses will be presented.

2.2 Results and discussion

Fig. 8 shows the temporal shapes of optically induced changes in THz pulse transmission measured on sample GaBiAs16 and GaBiAs35 for several optical pump pulse intensities. The rise time of the optically induced THz absorption transients indicating the temporal resolution of the experiment are of a similar order of magnitude (from 0.8 ps to 0.9 ps) and are determined by the THz pulse duration. The shapes of the decaying part of the transients are also similar – fast initial decay and slowly changing "tails" with 25 to 30 times smaller amplitude. Limited range of the optical delay times between the pump and the probe pulses did not allow us to determine the characteristic time of the slow decay component. On the other hand, characteristic time of the faster decay component is increasing when the sample's photoexcitation level is increased. Such a dynamics is typical when more mobile electrons are trapped faster than the

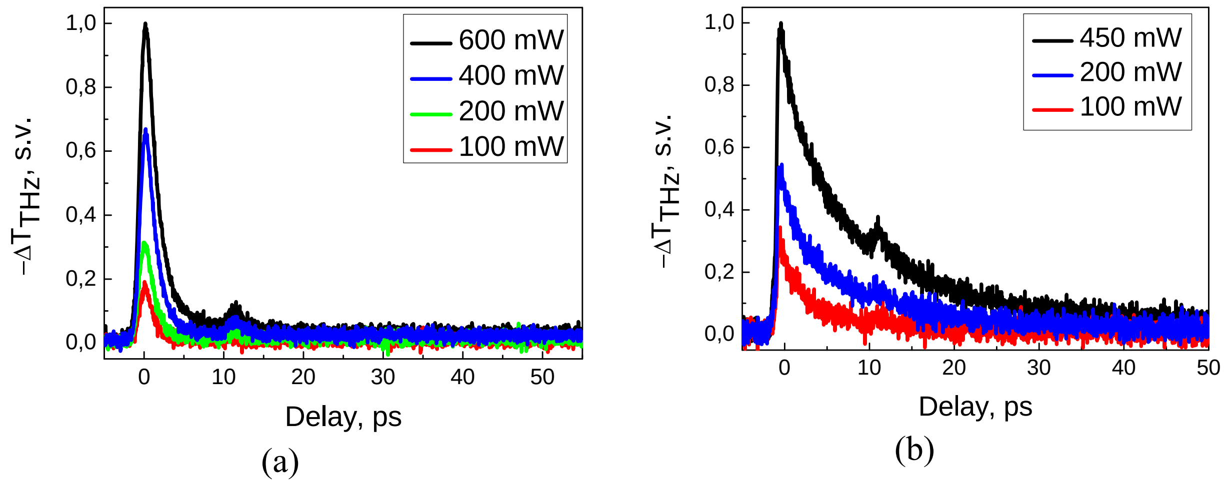


Fig. 8. Optically induced THz absorption transients measured for three different excitation levels (GaBiAs16 (a) and GaBiAs35 (b)).

photoexcited holes with a lower mobility and the electron traps become saturated with increasing excitation intensity.

This process was simulated by solving the rate equations for non-equilibrium electron dynamics in the conduction band and their capture at the trapping centers. Single trap model was used, and the trap emptying times has been assumed to be much longer than the electron capture time. The material parameters, electron trapping cross-section σ_n and the trap density N_f, were varied when fitting the model calculation with the experimental data. Fig. 9 shows the results of this fitting for three $GaBi_xAs_{1-x}$ layers. The best fit was obtained for $\sigma_n = 4.5 \cdot 10^{-13}$ cm^2 and $N_f = 4 \cdot 10^{16}$ cm^{-3} (samples GaBiAs16 and GaBiAs26) and $N_f = 3 \cdot 10^{16}$ cm^{-3} (sample GaBiAs35).

It has to be pointed out that obtained trapping cross-section value is larger than evaluated for As_{Ga} defects in LTG GaAs [19] and the trap densities in $GaBi_xAs_{1-x}$ are correspondingly smaller. We attribute it to more complete compensation of deep As_{Ga} donors, significant amount of which can be double-ionized.

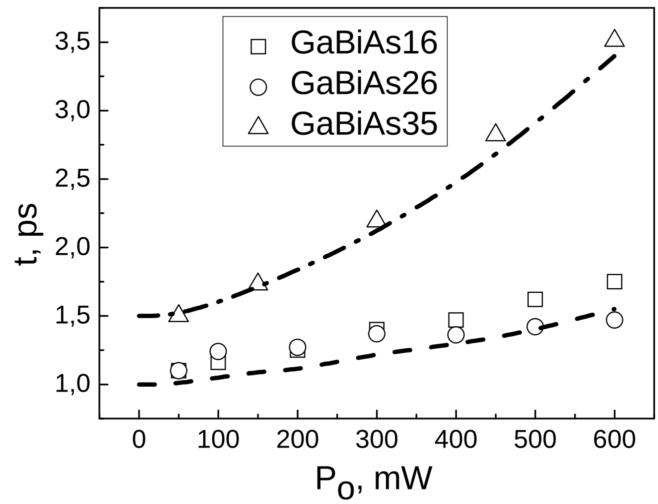


Fig. 9. Excitation level dependencies of the apparent electron lifetimes for three GaBiAs samples. Points - experiment, curves – modelling. Photoexcited carrier density at maximum average optical excitation power is equal to 9×10^{15} cm^{-3}.

These epitaxial GaBiAs layers were used for manufacturing photoconductive THz pulse detectors and emitters with integrated Hertzian dipole type antennas on their surfaces. The antennas had the shape of a coplanar transmission line with a 15 μm wide gap between Ti-Au electrodes that was used for the photoexcitation. For increasing the dark resistance of the device used as THz pulse emitter, GaBiAs layer was mesa-etched everywhere except for the photoconductive gap and the parts lying below the metal lines. The devices were equipped with hemispherical surface lenses made from high resistivity silicon; optically delayed parts of femtosecond Yb:KGW laser beam were used for their excitation.

Fig. 10 presents spectral content of THz pulses radiated and sampled by optoelectronic components manufactured from LTG GaBiAs layer. Average power lower than 20 mW was used for exciting the emitter and detector, average power of THz pulse beam as measured by a Golay cell was larger than 5 μW. It results in optical-to-THz power conversion efficiency of $\sim2\times10^{-4}$ which is better than for similar devices

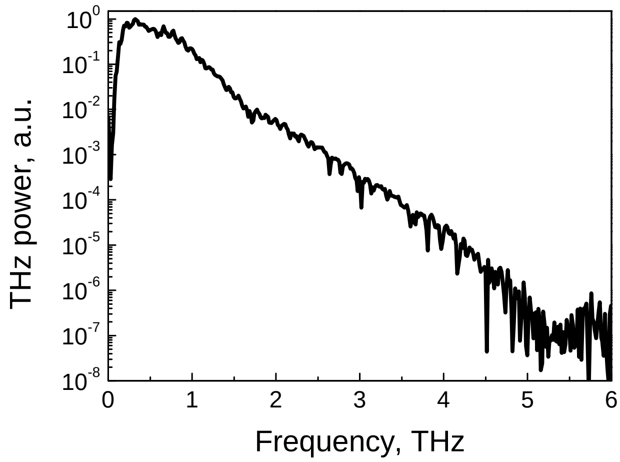


Fig. 10. Fast-Fourier-transform spectrum of THz pulse generated and detected by using components made from GaBiAs.

made from LTG GaAs and photoexcited by Ti:sapphire laser pulses.

The performance of these THz devices was later investigated by employing an Yb–doped fiber laser for their excitation. Fig. 11 shows THz pulses and their corresponding Fourier spectra for three different laser pulse durations, whereby the average optical power used for the excitation of THz emitter and for the detection was fixed at 8 mW. Fourier spectra

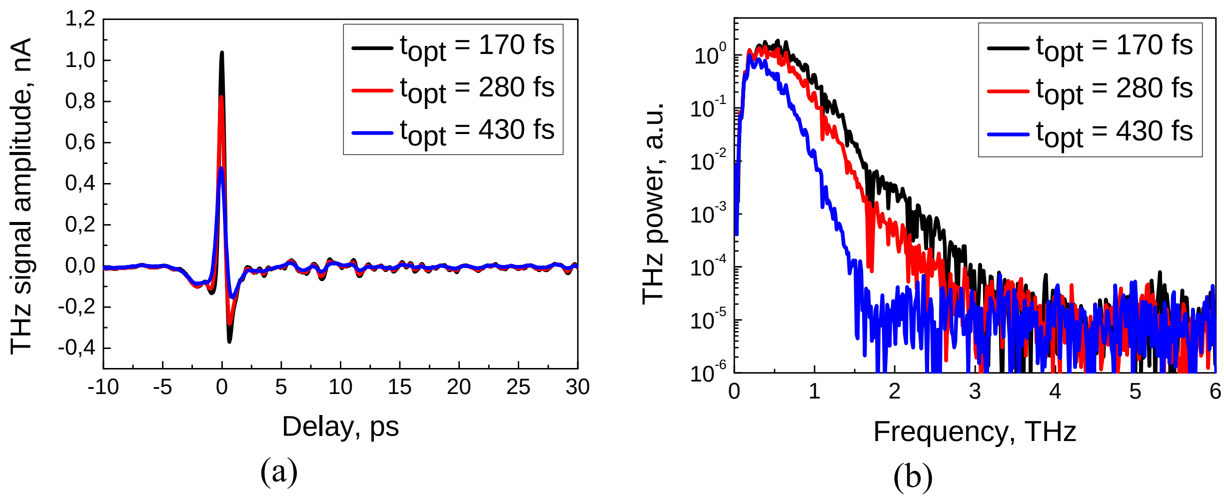


Fig. 11. (Color online) THz transients (a) and corresponding Fourier spectra (b) for three different laser pulse durations (indicated in the panels). Beams with average powers of 8 and 11 mW were used for the photoexcitation of the THz emitter and the detector, respectively.

obtained from the temporal THz profiles in the case of optimally compressed laser pulses (peak spectral intensity at 0.5 THz, bandwidth of 3.4 THz, and signal-to-noise ratio of 50 dB) from the fiber oscillator reveal a slightly worse performance as compared to the case of Yb:KGW laser with the same THz detector (see Fig. 10). This can be explained by a substantially shorter pulse duration and a larger peak pulse power of Yb:KGW laser. Both the amplitude and the bandwidth of the generated THz pulse decrease as the laser pulse duration increases.

2.3 Conclusions

GaBiAs epitaxial layer were grown by MBE technique at low substrate temperatures and investigated for their suitability in optoelectronic THz frequency range emitters and detectors activated by femtosecond near-IR laser pulses. Dark resistivity of the layers exceeding 2000 Ωcm and electron lifetimes of the order of 1 ps have allowed to use the devices made from this material in a time-domain spectroscopy system with a usable frequency range more than 4.5 THz and good optical-to-THz conversion efficiency.

Also we have described a compact THz-TDS system based on photoconductive antennae manufactured from GaBiAs layers and activated by low average power, 1030 nm femtosecond Yb-doped fiber oscillators The optical-to-THz power conversion efficiency of the GaBiAs emitters can exceed 5×10^{-4}, which is comparable with the best known pulsed THz sources. Obtained results reveal that sensitive ultrafast components from GaBiAs layers will facilitate the development of portable THz systems.

3. Terahertz time-domain spectroscopy system using a 1030 nm wavelength laser and photoconductive components made from low-temperature-grown GaAs (P6, P7)

In this section we will describe the manufacture of optoelectronic THz range components sensitive to near-infrared radiation from as-grown or annealed at moderate (~400°C) temperatures LTG GaAs layers. It is known that as-grown LTG GaAs contains a large density (10^{19} to 10^{20} cm^{-3}) As-antisite defects (As_{Ga}) that create an impurity band in the middle of the energy bandgap of GaAs. The optical absorption coefficient for the electron transitions from this band to the conduction band at room temperature is 10^4 cm^{-1} at 1 μm wavelength and 1.6×10^3 cm^{-1} at 1.55 μm [20]. The dark conductivity of as-grown LTG GaAs is dominated by electron hopping in the defect band, resulting in quite low electron mobility. On the other hand, the mobility of the photoexcited electrons in the conduction band should be comparable to that of the annealed material. Moreover, the breakdown field of the as-grown and moderately annealed layers is much larger than for layers annealed at higher temperatures (>600 °C) [21]. This property enables to apply large bias voltages to the emitters made from moderately annealed LTG GaAs, securing relatively large optical-to-THz radiation conversion efficiencies.

3.1 Experimental details

Epitaxial LTG GaAs layers were grown on (100)-oriented SI GaAs substrates by MBE at 250 ºC substrate temperature and a growth rate of ~1μm/h. During the growth, the layers were Be-doped in order to increase the number of ionized As_{Ga} defects [22]. Another LTG GaAs layer was grown under the same conditions on the top of AlAs/GaAs Bragg mirror with a maximum reflectance centered at 1μm wavelength. The thickness of all LTG GaAs layers was 1.4 μm. Both non-annealed and annealed layers were studied. The latter were annealed for 90s in a rapid thermal annealing oven at temperatures ranging from 400 ºC to 500 ºC. Coplanar Hertzian dipole antennas were manufactured from these layers with a photoconducting gap of 10-15 μm between the Ti-Au electrodes from LTG GaAs layers grown on a SI GaAs substrate. In order to collimate the emitted THz radiation, all antennas were equipped with hemispherical high resistivity Si lenses.

We used the oscillator from a high repetition rate femtosecond laser system Pharos (Light Conversion Ltd.) as an optical pulse source for THz pulse generation. The latter was based on a directly diode-pumped Yb:KGW crystal. Kerr-lens mode-locking was used to generate optical pulses with a pulse duration of 70 fs at wavelength of 1030 nm, spectral line-width (FWHM) of 22 nm and a pulse repetition rate of 76 MHz. The average output power of the oscillator was about 2 W. In the THz-TDS system, the laser beam was split into two parts of different intensity. The higher intensity beam (the power of ~25 mW) activated the photoconductive terahertz emitter and the lower intensity beam (the power of ~25 mW) activated the photoconductive THz pulse detector. The THz detector was made from $Ga_{0.96}Bi_{0.04}As$ layer, grown by MBE at low temperature (280 ºC) on a semi-insulating (100)-oriented GaAs substrate [23]. The electron trapping time in the detector material was shorter than 1 ps resulting in a Fourier transform spectra of the detected electromagnetic transients wider than 4 THz.

Additionally, for the optical pump – THz probe technique [24] 1.9 μm diameter pinhole was used to overlap the THz pulse and the photoexcitation beam on the sample. The spot size of the pump beam was larger than the diameter of the pinhole, so that the THz beam was sampling a nearly uniformly photoexcited region. To maximize the transmitted power through the pinhole, the THz beam was focused before and collimated after the pinhole by two hemispherical Teflon lenses. All measurements were performed at room temperature.

3.2 Results and discussion

3.2.1 Optical pump – THz probe experiments

The optical pump – THz probe experiment was performed on an as-grown LTG GaAs layer on a SI GaAs substrate and on a bare GaAs substrate. Curves representing the dependence of the transmitted THz signal versus time delay for the as-grown LTG GaAs (solid line) and for the bare GaAs substrate (dashed line) are shown in Fig. 12. The change of the transmitted THz signal amplitude, induced by the photoexcitation of the sample, is caused by free-carrier absorption at THz frequencies and is proportional to the induced photoelectron conductance. The experimental curve corresponding to the LTG GaAs layer shows fast dynamics immediately after the excitation with the laser pulse, but the shape of the transient is dominated by a slowly changing component. This component is also observed in the curve corresponding to the bare GaAs substrate and it is increasing linearly with the increase of the optical pump intensity. The slowly changing dynamics of the optically-induced THz absorption, obscuring the interpretation of pump-and-probe measurements, is probably caused by the electron excitation from various deep donor levels in the 600 μm thick SI GaAs wafer. Long rise time of the

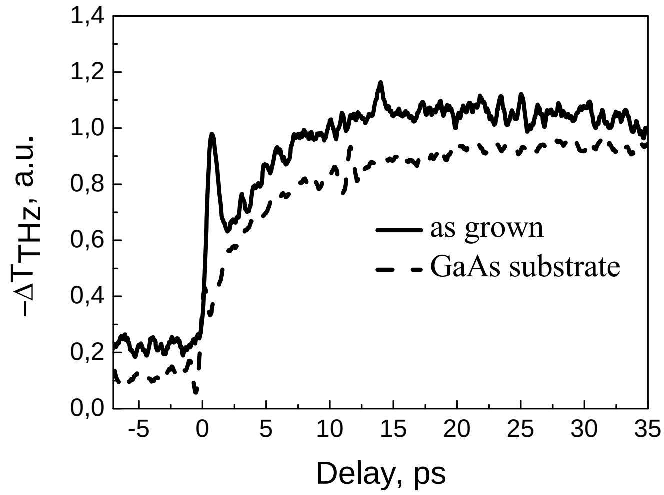


Fig. 12. Optical pump – THz probe measurements in an as-grown GaAs sample and in a bare GaAs wafer.

Substrate photoconductivity is, most probably, caused by the fact that 1.2 eV energy laser quanta excite the electrons with excess energies larger than the energy position of low mobility X valleys. Similar THz conductivity transients caused by slow inter-valley redistribution of the photoexcited electrons have been observed before in germanium [25] and in several narrow-gap semiconductors [26].

To eliminate the effect of the GaAs substrate, the same experiment was repeated on the LTG GaAs layer grown on a Bragg reflector. Fig. 13 shows the shape of the optically induced THz absorption transient for an as-grown LTG GaAs sample. As it

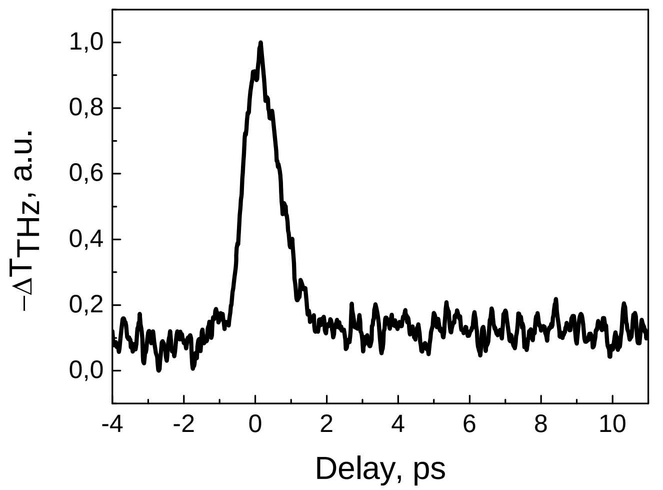


Fig. 13. Optically induced THz absorption transients measured for the LTG GaAs layer grown on the Bragg reflector.

can be seen, the introduction of the Bragg reflector into the structure has completely removed the slowly changing transient component as well as the constant background signal. The FWHM duration of the induced THz absorption transient is ~1 ps and its rise and fall-times are similar. Thus, we can conclude that the electron lifetime in the layer is shorter than the temporal resolution of the experiment that is determined by the duration of the THz pulses (~0.8 ps).

We examined the induced THz absorption dependence on the near-infrared excitation level for three samples. The as-grown and annealed at 420 ºC and 450 ºC temperatures LTG GaAs samples were grown on Bragg reflector. The experimental dependence for the as-grown sample is shown in Fig. 14 and is marked by squares. The experimental curves corresponding to the samples annealed at the temperatures of 420 ºC and 450 ºC are also shown in Fig. 14 and are marked by circles and triangles, respectively.

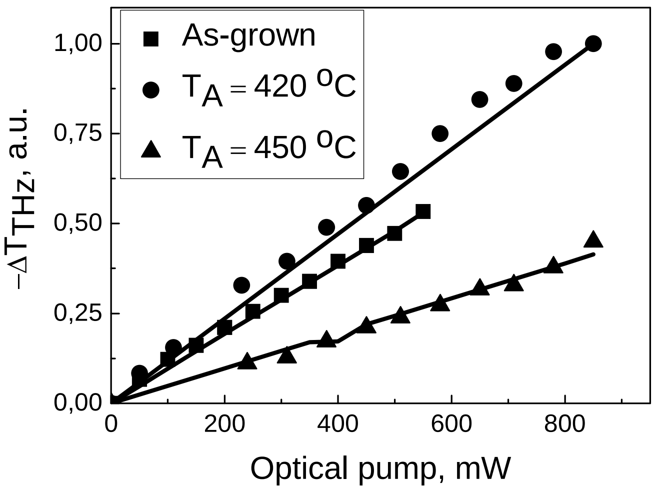


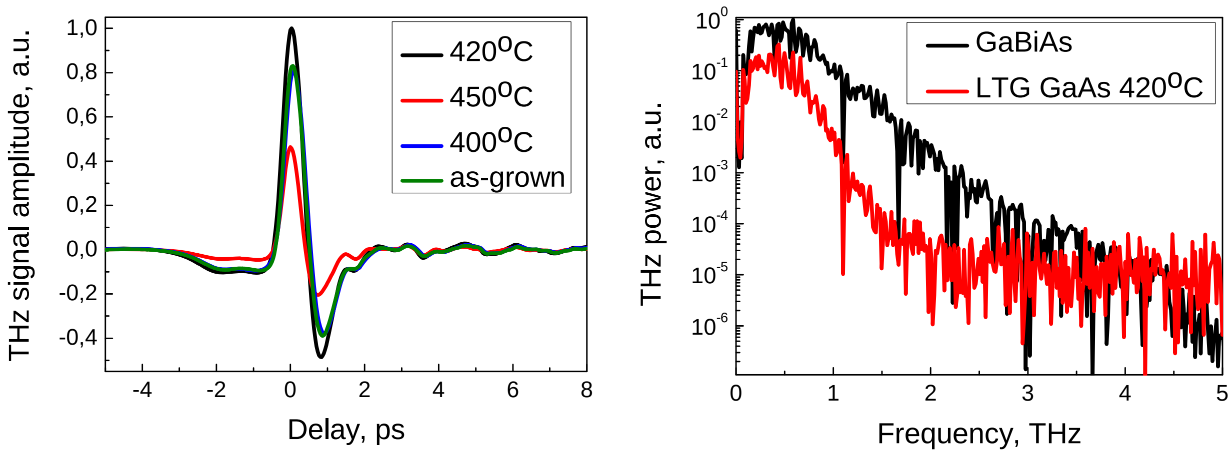
Fig. 14. Dependence of the THz transmittance peak value on average laser pulse power.

As it can be seen, THz absorption in all the studied samples increases linearly with the increment of photoexcitation level. This fact provides strong evidence that in the as-grown and moderately annealed LTG GaAs the photo-absorption at 1 μm wavelength is dominated by the electron transitions from the As-related defects rather than by two-photon or two-step absorption processes.

The largest THz absorption signal was obtained from the sample annealed at 420°C. It was slightly lower for the as-grown sample and approximately three times lower for the sample annealed at 450°C.

3.2.2. THz emitter and THz detector characterization

In this section, photoconductive switches manufactured from the materials studied in the former experiment will be described. For comparison, in the experiments we investigated additional photoconductive switches made from LTG GaAs annealed at 400 °C, 500 °C and higher temperatures. All the studied samples were grown without introduction of a Bragg reflector. All the switches were tested as the THz pulse emitters in the THz-TDS system, with the photoconductive detector made from the GaBiAs layer and illuminated with the average laser power of 20 mW for the THz pulse sampling. Experimental curves corresponding to the THz pulses emitted by these switches are shown in Fig. 15a.



(a) (b)

Fig. 15. THz pulses generated by photoconductive emitters made from as-grown and annealed at various temperatures LTG GaAs and sampled by the GaBiAs detector (a). Fourier spectrum of the THz pulse generated by LTG GaAs photoconductor annealed at 420°C and measured by the detectors made from the same material and from the epitaxial GaBiAs layer (b).

THz pulses emitted by the switches made from as-grown and annealed at 400 °C temperature samples were almost identical. The THz pulse with the largest amplitude was emitted by the device made from the layer annealed at the temperature of 420°C (solid line) on Fig. 15a. For this layer, the optically induced THz absorption was also the largest, as it was shown in Fig. 14. Photoconductive antennas made from the as-grown and the annealed at 450°C material emitted slightly lower-amplitude THz pulses. No THz pulses were detected from the devices annealed at 500°C and higher temperatures.

In the second part of this experiment, we compared the performance of GaBiAs detector to that of the detector based on the LTG GaAs, which was annealed at 420 C. The signal generated by the most efficient emitter (LTG GaAs annealed at 420 C) was detected by each of the detectors mentioned above. The Fourier transform (FT) spectra of those signals are shown in Fig. 15b. The FT spectrum of the signal detected by the GaBiAs detector (solid line) has useful bandwidth reaching to 3 THz and its signal-to-noise ratio exceeds 50 dB. The FT spectrum corresponding to the photoconductive switch manufactured from the LTG GaAs (dashed line) is almost two times narrower and its sensitivity is ~15 times lower comparing to the GaBiAs detector.

Most distinctive property of the photoconductive THz emitters manufactured from LTG GaAs and annealed at moderate temperatures, is their high breakdown voltage. An alternating meander-shaped voltage waveform with the amplitude varying between -150 V and +150 V was applied to a 10 μm wide photoconducting gap. The dependence of the THz pulse amplitude generated by the device annealed at 420°C on the bias voltage is shown in Fig. 16.

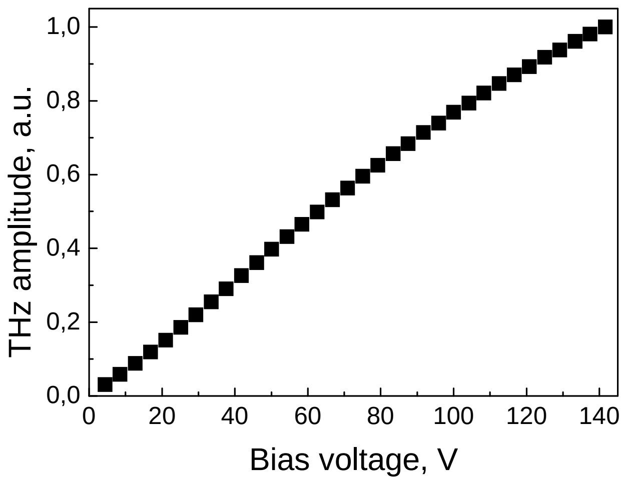


Fig. 16. Normalized THz signal amplitude generated by the terahertz emitter, annealed at 420 ºC, versus bias voltage.

The THz pulse amplitude increases linearly with the increasing voltage. Thus, we can conclude that the electric breakdown field of the gap is larger than 150 kV/cm. This value is much higher than the value of the average electrical breakdown field of the LTG

GaAs annealed at high temperatures, which is typically around 50kV/cm [21]. A significant increase of this parameter for as-grown and annealed at lower temperatures devices could be explained by the influence of the hopping conduction. The hopping current prevents the nucleation of the high-field domain in the anode region and leads to a more homogeneous electrical field distribution along the bandgap. Thus, the avalanche breakdown in the device occurs at higher voltage [21].

3.3 Conclusion

In conclusion, photoconductive antenna manufactured from as-grown and moderately annealed LTG GaAs layers activated by 1030 nm wavelength femtosecond duration laser pulses were used for terahertz pulse generation and detection. An optical pump – terahertz probe technique was used to study the ultrafast photoconductivity in these layers. It has been shown that this photoconductivity caused by the electrons excited from deep As_{Ga} donor levels is the largest for LTG GaAs annealed at the temperature of about 420 °C. In the as-grown material it is lower because of the lower electron mobility, whereas higher annealing temperatures are leading to a significant reduction of the As_{Ga} defects.

As GaAs has a much larger energy bandgap than other semiconductors used for manufacturing optoelectronic THz components sensitive to 1 μm wavelength laser radiation, the investigated THz emitters could be biased to relatively large voltages. Because of their high optical-to-THz radiation conversion efficiencies and reliability, the THz emitters made from moderately annealed LTG GaAs are preferable choice for THz-TDS systems activated with femtosecond near-infrared lasers. On the other hand, the sensitivity of the pulsed THz detectors made from this material is by an order of magnitude worse than for GaBiAs detectors – the best detecting devices for THz-TDS systems using the lasers of the near-IR spectral range.

Santrauka

Disertacijos darbo tikslas buvo sukurti ir ištirti puslaidininkinius terahercinių (THz) impulsų emiterius ir detektorius, skirtus sistemoms, naudojančioms 1 μm bangos ilgio femtosekundinę lazerinę spinduliuotę.

THz impulsų generavimo ir detektavimo sistema, kurios optoelektroninius puslaidininkinius komponentus aktyvuoja femtosekundiniai lazerio impulsai, yra plačiai taikoma terahercinėje laikinės srities spektroskopijoje. Tradiciškai tokiose sistemose naudojami Ti:safyre femtosekundiniai lazeriai, kurių spinduliuotės bangos ilgis yra ~800 nm. Šios sistemos nėra patogios dėl jų matmenų, nes lazeriai turi sudėtingą kelių pakopų kaupinimo sistemą.

Pastaruoju metu THz impulsų generavimui vis dažniau naudojami femtosekundiniai kietakūniai ir šviesolaidiniai lazeriai, kurių spinduliuotės bangos ilgis patenka į artimosios IR spinduliuotės sritį. Tačiau šios sistemos vis dar neturi tinkamos medžiagos fotolaidiems elementams gaminti, kurie būtų žadinami 1 – 1,55 μm bangos ilgio lazeriais. Tokios medžiagos, visų pirmą, turi būti jautrios optinei spinduliuotei, o jų draustinės energijos tarpas turi atitikti žadinamos spinduliuotės fotonų energiją, be to sluoksniai turi pasižymėti didele tamsine varža bei labai trumpomis krūvininkų gyvavimo trukmėmis (~ 1 ps).

Šioje disertacijoje yra pateikiami THz impulsų generavimo panaudojus puslaidininkių paviršius ir fotolaidžias antenas rezultatai, žadinant 1 μm bangos ilgio femtosekundiniais lazerio impulsais.

Darbe buvo tirti InAs, InSb, CdHgTe, Ge, GaBiAs, InGaAs, GaAs, HgMnTe, InP, InN paviršiniai emiteriai, įvairūs GaBiAs, ŽT InGaAs detektoriai bei ŽT GaAs emiteriai. Parodyta, kad geriausias paviršinis emiteris 1 μm bangos ilgio lazerinei spinduliuotei yra p-InAs kristalas. Be to nustatyta, kad žadinant šia lazerine spinduliuote InAs kristalus sugeneruotam THz signalui didžiausią įtaką turi elektriniu lauku indukuoto optinio lyginimo (EFIOR) mechanizmas, o aprašant InSb kristale sugeneruotą THz signalą reikia atsižvelgti į abu netiesinius optinius mechanizmus: optinį lyginimą ir EFIOR.

Geriausi THz detektoriai minėtajam žadinimo lazerio bangos ilgių diapazonui yra ŽT GaBiAs. Juose elektronai turi didelį judrį bei trumpas gyvavimo trukmes, kas lemia jų geresnį jautrumą THz spinduliuotei bei platesnę dažnių juostą.

Taip pat buvo parodyta, kad iš ŽT GaAs pagamintus emiterius, galima naudoti ir su 1 μm bangos ilgio lazerine spinduliuote, tačiau juos reikia atkaitinti žemesnėse nei 500 °C temperatūrose. Tada emiterių charakteristikas lems optinė sugertis iš arseno pakaitinių atomų lygmenų ir elektronų pagavimas juose.

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List of publications related to the summary of doctoral dissertation

Papers

P1. V. Pačebutas, **A. Bičiūnas**, K. Bertulis, and A. Krotkus, „Optoelectronic terahertz radiation system based on femtosecond 1 µm laser pulses and GaBiAs detector“, Electron. Lett., **44**, 19 (2008).

P2. V. L. Malevich, A. Krotkus, **A. Bičiūnas**, and V. Pačebutas, „Terahertz emission from femtosecond laser illuminated (112) surfaces of InSb“, J. Appl. Phys., **104**, 113117 (2008).

P3. **A. Bičiūnas**, V. Pačebutas, A. Krotkus, „Terahertz pulse emission from semiconductor surfaces illuminated by femtosecond Yb:KGW laser pulses“, Physica B, **404**, p. 3386-3390 (2009).

P4. V. Pačebutas, K. Bertulis, **A. Bičiūnas**, A. Krotkus, „Low-temperature MBE-grown GaBiAs layers for terahertz optoelectronic devices“, Phys. Stat. Sol. C, **6**, p. 2649-2651 (2009).

P5. V. Pačebutas, **A. Bičiūnas**, S. Balakauskas, A. Krotkus, G. Andriukaitis, D. Lorenc, A. Pugžlys, and A. Baltuška, „Terahertz time-domain-spectroscopy system based on femtosecond Yb:fiber laser and GaBiAs photoconducting components“, Appl. Phys. Lett., **97**, 031111 (2010).

P6. **A. Bičiūnas**, A. Geižutis and A. Krotkus, „Teraherzt generation by photoconductors made from low-temperature-grown GaAs annealed at moderate temperatures“, Electron. Lett., **47**, 2 (2011).

P7. **A. Bičiūnas**, J. Adamonis and A. Krotkus, „Terahertz time-domain-spectroscopy system using a 1 micron wavelength laser and photoconductive components made from low-temperature-grown GaAs“, J. of Infrared Milli. Terahz. Waves, **33**, p. 183-191 (2012).

Conference contributions

K1. V. Pačebutas, K. Bertulis, **A. Bičiūnas**, A. Krotkus, „GaBiAs layers for terahertz optoelectronics devices activated by 1 µm wavelength laser pulses“, The

European Materials Research Society (E-MRS) 2009 Fall Meeting, 14-18 September 2009, Warsaw, Poland.

K2. V. Pačebutas, K. Bertulis, **A. Bičiūnas**, A. Krotkus, „Low-temperature MBE-grown GaBiAs layers for terahertz optoelectronic devices“, 15th Semiconducting and Insulating Materials Conference (SIMC XV), 15-19 June 2009, Vilnius, Lithuania.

K3. V. Pačebutas, **A. Bičiūnas**, K. Bertulis, and A. Krotkus, „GaBiAs layers for terahertz optoelectronic devices activated by 1 μm wavelength laser pulses“, XXXIX “Jaszowiec” International School & Conference on the Physics of Semiconductors, 19-24 June 2010, Krynica-Zdroj, Poland.

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