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Terahertz excitation spectroscopy of Bi/GaAs heterostructures

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ABSTRACT

In this work, heterojunctions consisting of thin monoelemental bismuth (Bi) layers have been grown on crystalline GaAs substrates with various doping types and investigated using THz pulse generation excited via femtosecond optical pulses of different wavelengths. It was determined that the influence of the layers of Bi can be approached in two distinct ways: first, through the additional absorption of the optical pulse and second, through changes in the electron energy band line-ups at the heterojunction. The strongest influence of the latter effect was observed at the interface between Bi and p-GaAs when the internal electric field exceeded the saturation field of the drift velocity of the p-GaAs holes. Such high fields can lead to an overshooting effect of the holes when the holes move at speeds above 2×10^7 cm/s and emit a much stronger THz signal than in the case of p-GaAs. This finding is promising for the development of efficient surface THz pulse emitters based on p-GaAs and other p-type semiconductors.

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I. INTRODUCTION

Two-dimensional (2D) materials are ultra-thin structures with atomic or molecular thicknesses, enabling unique electronic, optical, and mechanical properties due to quantum confinement and high surface-to-volume ratios. These materials, including graphene¹ and transition metal dichalcogenides (TMDs),² have profoundly impacted various fields, such as electronics, photonics, and energy storage, by enabling novel functionalities at the nanoscale. Recent research has expanded to include other atomically thin single-element materials, such as germanene,³ silicene,⁴ and tellurene,⁵ as well as group V elements, including phosphine,⁶ arsenene,⁷ antimonene,⁸ and bismuthene.⁹ Interest in thin bismuth (Bi) layers has recently grown, as the potential of the few-atomic-layer structures of this material to act as topological insulators has become evident.^{10,11} In addition, nanometer-thin bismuth layers are being investigated for a variety of applications, including sensors,¹² thermoelectricity,¹³ contacts for Na-ion batteries,¹⁴ and femtosecond optical switches.¹⁵

Bulk bismuth crystals are semi-metallic, with anisotropic electron energy dispersion and low carrier effective masses. When a Bi

layer is thinned to approximately 30 nm or below, it demonstrates semiconductor properties instead of semimetal characteristics. This conversion happens due to strong confinement effects in electronic states of the material.¹⁶ Bandgap tunability has been observed at further thinning of Bi layers,¹⁷ and samples with only a few atomic layers thick grown on SiC exhibited an energy gap reaching 0.8 eV.¹⁸ In another study, when bismuthene was grown on Ag (111), an even larger topological gap of 1 eV was observed.¹⁹ So far, the growth of high quality crystalline thin layers of Bi has been achieved only on InAs,²⁰ InP,²¹ and, most recently, on (111) oriented silicon substrates.^{22,23} However, growth on a (111) GaAs substrate resulted in a honeycomb structure of a Bi monolayer with a larger lattice constant than in previous works.²⁴ It is critically important to determine the electrical and crystalline properties of bismuthene layers, as well as to identify new substrates that can support high quality crystal growth.

Ultrashort electrical pulses generated employing femtosecond laser light are now extensively used to probe the physical characteristics of various materials, including semiconductors. These sub-picosecond electromagnetic pulses are applied in techniques, such

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as THz time-domain spectroscopy (TDS), optical-pump THz-probe (OTTP) measurements, as well as for the THz excitation spectroscopy (TES).²⁵ TES has been used to extract electron energy band structure parameters, such as a subsidiary valley position²⁶ and heterostructure band offsets²⁷ by analyzing how the amplitude of the emitted THz pulse depends on the energy of the exciting photons. This technique has recently been used to investigate several layered materials, including GaSe²⁸ and transitional metal dichalcogenides (TMD).²⁹ The mobility and lifetimes of charge carriers in thin tellurium layers have also been determined using this method.³⁰

It has been shown that, when excited with femtosecond laser pulses, both bulk bismuth crystals³¹ and polycrystalline bismuth layers with thickness ranging from 30 to 100 nanometers³² emit electromagnetic pulses in the THz range. According to another study,³³ the nonlinear Hall effect observed in Bi films at room temperature results in efficient frequency doubling and tripling in the terahertz range. Ultrathin bismuth films deposited on (111) silicon have also demonstrated efficient terahertz (THz) generation, which arises due to uncompensated lateral photocurrents caused by diffusive electron scattering at the Bi/Si interface.³⁴ Furthermore, TES measurements revealed that the direct bandgap increases from 0.25 to 0.5 eV as the thickness of the crystalline bismuth layer is reduced from 30 to 7 nm.³⁵

In the present work, thin Bi layers were grown on (100) oriented GaAs substrates using molecular beam epitaxy (MBE). The growth of such layers on this specific lattice orientation of GaAs has not been previously reported. THz emission was observed from this structure, and THz pulse excitation spectra were measured. It was shown that THz emission from the Bi/GaAs heterostructure, when excited at different optical wavelengths, can arise from two physical mechanisms.

II. LAYER GROWTH AND EXPERIMENTAL DETAILS

Bi samples were grown on (100) oriented GaAs substrates with different doping using a Veeco GENxplor MBE system equipped with a conventional dual-filament bismuth source. The substrate temperature was controlled by a thermocouple. Before growth, the sample temperature was raised to 600 °C in an arsenic flux and maintained for ~10 min until arsenic oxide desorbed. After desorption, the substrate temperature was lowered to 50 °C and a bismuth layer was grown. The growth rate was set to ~36 nm/h. X-ray diffraction (XRD) measurements (Fig. S1 in the supplementary material) indicated the presence of crystalline bismuth layers and revealed that its orientation corresponds to the 003 direction, assuming a hexagonal lattice.

THz pulse excitation spectroscopy involves a THz time-domain spectroscopy (THz-TDS) setup where the sample under investigation serves as the THz emitter. The THz-TDS system employed in this study utilized a Yb:KGW laser source with a central wavelength of $\lambda \approx 1030$ nm, a pulse duration of 180 fs, and a pulse repetition rate of 200 kHz (PHAROS Laser Light Conversion Ltd., Vilnius, Lithuania). A small part of the laser beam power, about 5 mW of average power, was separated via a beam splitter and used to illuminate the GaAsBi photoconductive antenna detector (Teravil Ltd.). The main part of the laser beam

was directed to an optical parametric amplifier (OPA) ORPHEUS (Light Conversion Ltd.), enabling wavelength tuning of the femtosecond pulses from 640 nm to about 2600 nm without pulse duration change. The bismuth samples were excited using a p-polarized pump beam with a collimated beam of 2 mm diameter (at 1/e² FWHM). The beam was incident at a 45° angle, and measurement was performed in reflection geometry.

III. RESULTS AND DISCUSSION

Heterostructures consisting of 9 and 30 nm thick bismuth layers grown by MBE on three (100) GaAs substrates, SI-GaAs, n-GaAs ($n = 2 \times 10^{18} \text{ cm}^{-3}$), and p-GaAs ($p = 1 \times 10^{17} \text{ cm}^{-3}$), were fabricated and investigated. Terahertz excitation spectra (TES) of the heterostructures with a 9 nm-thick Bi layer on each of the substrates are shown in Fig. 1. The spectra were measured in the reflection geometry by illuminating the Bi-side surfaces at a 45° angle with p-polarization at a fixed average power of 20 mW, but at different photon energies. For convenient comparison, all the results presented below were normalized to the same number of photons incident on the surface of the structure.

These spectra are substantially different. Their common feature is weak THz emission when the photon energy is below the GaAs bandgap of $\epsilon_g = 1.42$ eV. The THz amplitude dependence on the photon energies above ϵ_g becomes stronger and substantially different for Bi layers on different substrates. The opposite polarities of THz pulses generated from samples on n-type and p-type substrates can be explained by oppositely oriented internal electric fields at the GaAs and Bi layer interface, creating reversely directed photocurrent transients. Meanwhile, the observed polarity change of the THz pulse near photon energies of $h\nu \approx 1.7$ eV is more

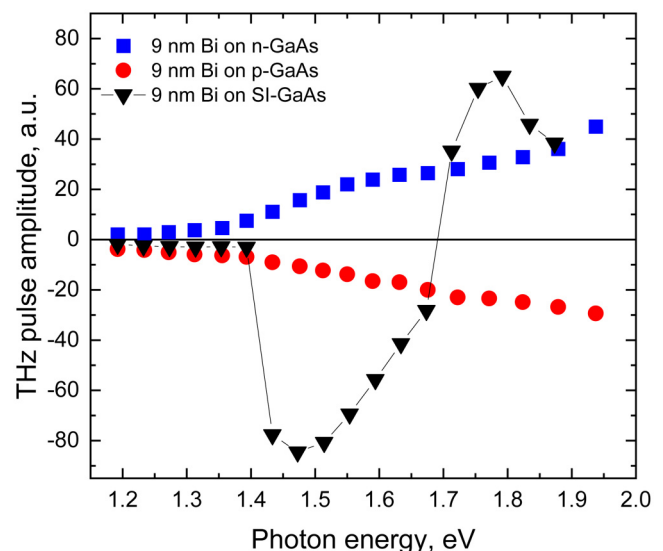


FIG. 1. THz excitation spectra of heterostructures containing 9 nm-thick Bi layers deposited on SI-GaAs, p-GaAs, and n-GaAs substrates, as indicated. The lines are guides to the eye.

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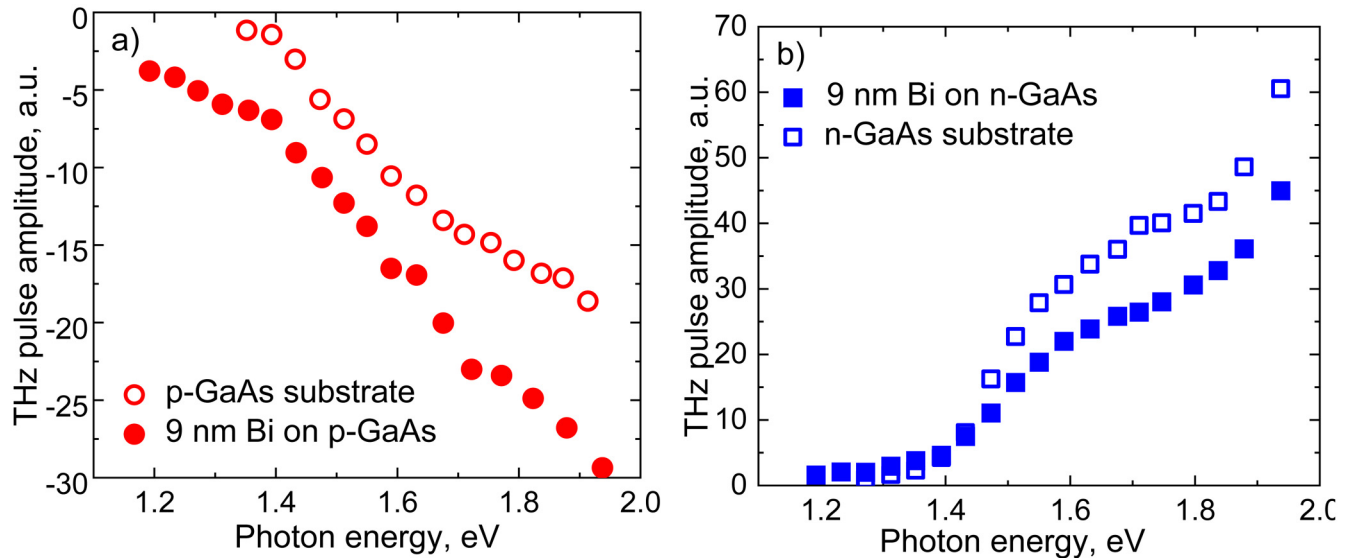


FIG. 2. THz excitation spectra for Bi/GaAs heterostructures grown on p-GaAs (a) and n-GaAs (b) substrates. The spectra measured on corresponding bare substrates are provided for comparison.

challenging to interpret. This phenomenon will be addressed in more detail below.

Figure 2 compares the TES measured on Bi heterostructures grown on doped GaAs substrates with the spectra of the corresponding bare substrates. For both substrates, a significant increase in the THz pulse amplitude is observed only when the photon energy exceeds the GaAs bandgap. This is an indication that the THz emission originates from the mechanism in GaAs substrates. Therefore, assuming that the THz amplitude depends linearly on the laser excitation power, the optical absorption coefficient of the bismuth layer can be estimated by comparing the THz pulses emitted from the heterostructure and from the corresponding substrate. In the photon energy range between 1.5 and 1.7 eV, the absorption coefficient in the bismuth layer is found to be within the range between 3×10^5 and $4 \times 10^5 \text{ cm}^{-1}$,³⁶ which is more than an order of magnitude higher than that of GaAs (1×10^4 to $3 \times 10^4 \text{ cm}^{-1}$) in this spectral region.³⁷

While the difference in TES characteristics observed for Bi heterojunctions on heavily doped n-GaAs substrates compared to the corresponding bare substrates can be explained by an absorption mechanism of the excitation laser pulse in the Bi layer, the case of Bi on SI-GaAs and on p-GaAs heterostructures is completely different. For the p-GaAs substrate, the THz emission from the heterostructure is stronger than that from the bare substrate. In contrast, for the heterostructure grown on SI-GaAs, a reduction in the amplitude of THz pulses is observed when the sample is excited through the Bi layer, along with a reversal in the polarity of these pulses. This effect is illustrated in Fig. 3, which shows THz pulses generated on an SI-GaAs substrate and in two heterojunctions with Bi layers of different thicknesses, illuminated by femtosecond optical pulses

with photon energies close to the negative and positive extrema of the TES spectrum.

It should be noted that in Fig. 3(b), the amplitude of the pulses emitted from the substrate is reduced by a factor of 5, whereas the amplitudes of the pulses emitted from the heterostructures are shown at their actual scales. The comparison of these pulses reveals that the amplitude at the negative extreme of TES is approximately by a factor of two larger than that at the positive extreme. Also, the THz pulse amplitude of the thinner sample is several times higher since reflectivity increases as a result of semi-metallic nature of the sample. Moreover, at higher photon energies, the difference between the pulses generated from the heterostructure and the substrate becomes significantly greater than in the samples grown on heavily doped GaAs substrates. These observations imply the presence of an additional THz emission mechanism in the Bi/SI GaAs heterostructure. The mechanism generates signals of the opposite polarity compared to those produced by conventional mechanisms. The nature of this mechanism was investigated by studying the energy band alignment in Bi/GaAs heterostructures.

The energy band line-ups of the heterostructures consisting of Bi layers grown on different GaAs substrates were simulated using Silvaco software. The bandgap energy of the 9 nm thick bismuth layer was set to 0.1 eV,³⁸ and the electron affinity (χ) was assumed to be 4.0 eV;³⁹ the value of the GaAs bandgap was taken to be 1.42 eV, regardless of doping. The results of this simulation are presented in Fig. 4. Furthermore, Fig. 5 illustrates the spatial distribution of the internal electric field arising near the heterojunctions. These fields play a significant role in shaping the properties of the THz emission observed in various heterostructures.

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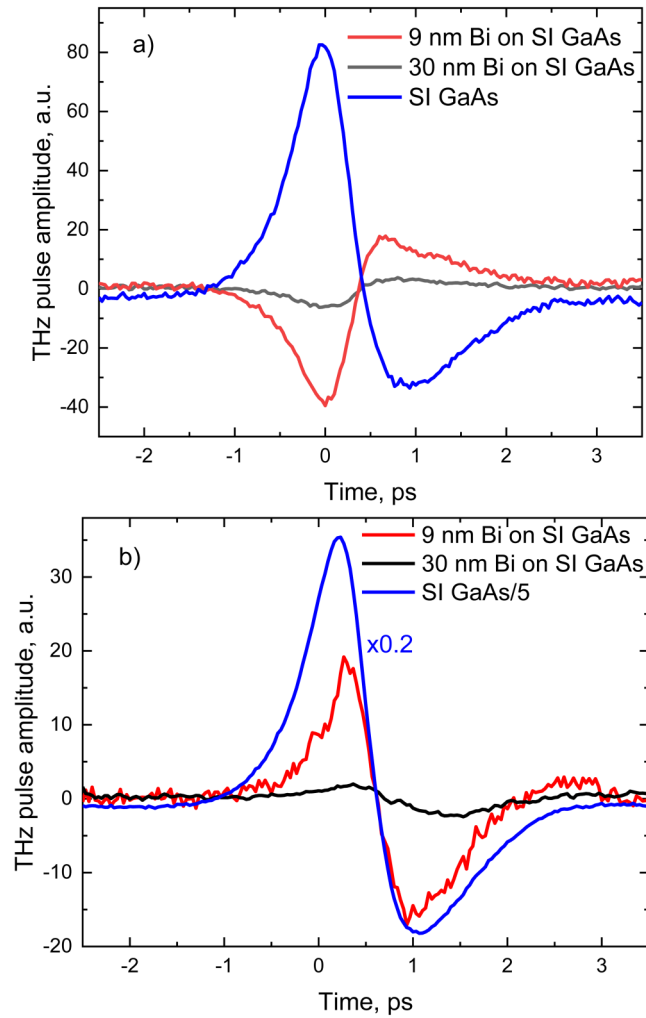


FIG. 3. THz pulses from Bi/GaAs heterostructures containing Bi layers of two different thicknesses and from a bare SI-GaAs substrate radiated at femtosecond pulses of photon energies of 1.46 (a) and 1.79 eV (b).

In a Bi/n-GaAs structure, the internal electric field exists only in the immediate vicinity of the junction. Although its polarity is directed to accelerate photoexcited electrons, the overall signal generated in this structure is smaller than that in the structure without a Bi layer, suggesting that the optical absorption in the Bi layer is even greater than previously estimated. The strongest internal electric field was calculated in the junction between Bi and p-GaAs. It is above the hole drift velocity saturation field (~ 100 kV/cm for p-GaAs⁴⁰) within the depth of 100 nm from the surface. Furthermore, as shown by the Monte Carlo simulations of hole transport in GaAs performed in the same study,⁴⁰ such high fields can lead to a hole velocity overshoot effect. As a result, the photoexcited holes can move at velocities greater than 2×10^7 cm/s even over the distances of 100 nm, leading to a larger THz signal than in the case of a bare p-GaAs substrate.

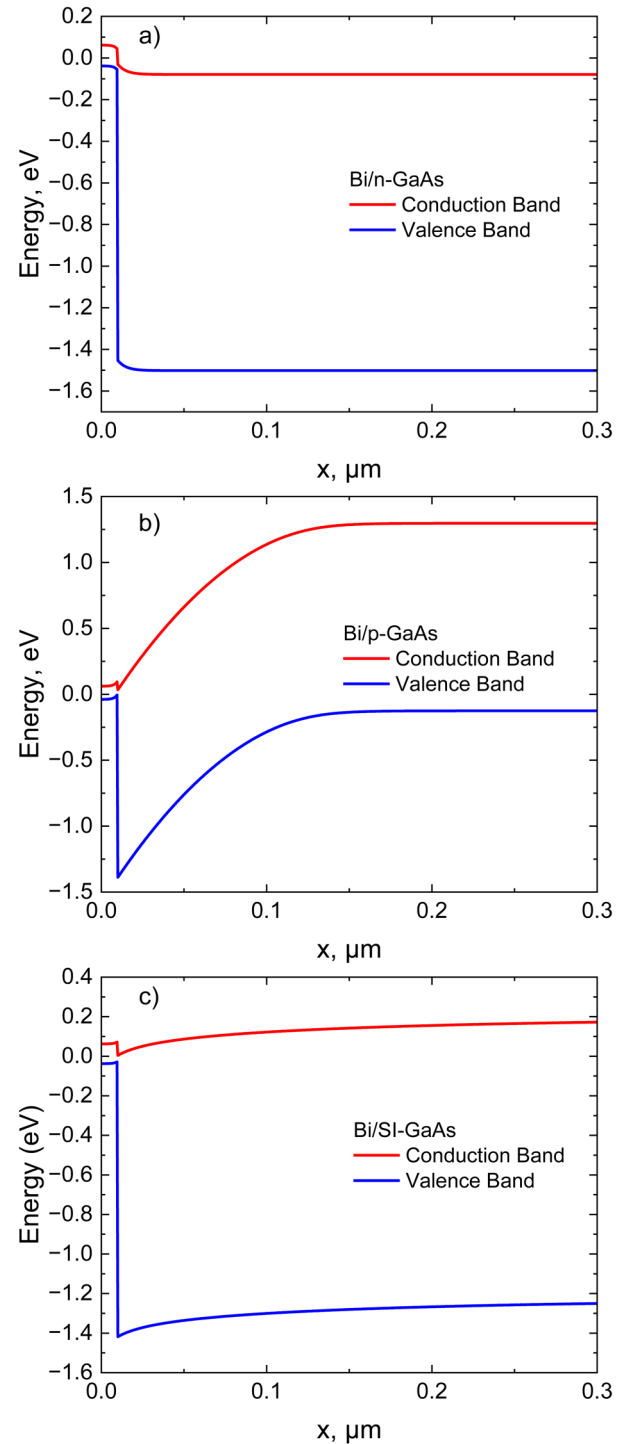


FIG. 4. The energy band line-ups of (a) Bi/n-GaAs, (b) Bi/p-GaAs, and (c) Bi/SI-GaAs heterostructures. The bandgap energy of the 9 nm thick bismuth layer was set to 0.1 eV, and the electron affinity (χ) was assumed to be 4.0 eV, while the GaAs bandgap was set to 1.42 eV, regardless of doping.

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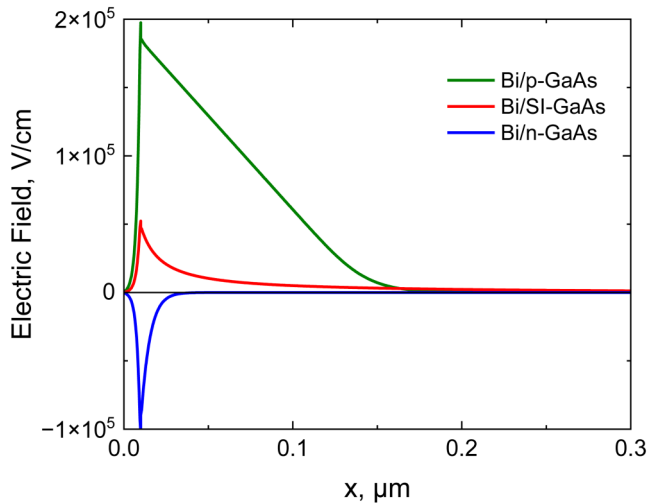


FIG. 5. The internal electric field distribution in the vicinity of Bi/GaAs heterojunctions. The same parameters were used in calculations as in Fig. 4.

Although the internal electric field in the Bi/Sl-GaAs heterojunction is relatively weak, it extends deep into the GaAs crystal due to the large Debye screening length. When the photon energy only slightly exceeds the GaAs bandgap, the absorption length is also large, meaning that most photoexcited charge carriers are influenced by this field. In semi-insulating crystals, where the surface electric field is generally weaker compared to that in heavily doped materials, two physical mechanisms associated with the spatial separation of electrons and holes dominate the THz pulse emission: the faster diffusion of electrons into the volume (the photo-Dember effect)⁴¹ and a fast ballistic motion of electrons in the same direction.⁴² Both mechanisms become more efficient at higher photon energies when the velocities of excited electrons and the gradients in the photo-excited carrier density are larger. At lower photon energies, the influence of the internal electric field direction would become apparent. Simulations of the band structure presented in Fig. 4 demonstrate that both Bi/Sl-GaAs and Bi/p-GaAs heterojunctions exhibit band bending in the same direction, resulting in an intrinsic electric field oriented similarly in both cases. Correspondingly, the experimental spectra shown in Fig. 1 reveal that the THz pulse amplitude maintains the same polarity when excited at photon energies just above the GaAs bandgap (in the range of 1.4–1.6 eV). In this case, the electrons would move opposite to the direction of the field toward the heterojunction. Electric field occurring due to a photo-Dember effect is of opposite direction compared to an intrinsic field in a p-type semiconductor. This would force electrons to move in the opposite direction, toward the GaAs bulk and result in a dynamically varying photocurrent, thus giving rise to a THz pulse of opposite polarity. This provides a possible explanation for the reversal observed in the Bi/Sl-GaAs heterojunction of Fig. 1 when exciting with a photon energy of 1.7 eV and higher.

IV. CONCLUSIONS

For the first time, heterojunctions composed of thin monoelemental bismuth layers were grown on crystalline GaAs substrates with n and p doping types. The band structure of these heterojunctions was investigated by studying THz radiation generated by excitation with femtosecond optical pulses at various wavelengths.

The results for heterostructures grown on differently doped substrates showed significant differences. In all structures studied, the THz radiation pulses originated from physical processes within the GaAs substrate. The presence of the Bi layers influenced the emission in two ways: through additional absorption of the optical pulse within the Bi layer and by altering the electron energy band line-ups at the heterojunction. The Bi layer exhibited THz pulse emission when excited with photons below the GaAs bandgap; however, the emission intensity was notably lower compared to that observed with excitation above the GaAs bandgap.

The THz excitation spectra of the heterostructures grown on n-GaAs substrates showed the smallest deviations from those of the bare substrate. These changes can be explained by a reduction in the power of the incident exciting optical pulse due to additional absorption in the Bi layer. The estimated absorption coefficient at a photon energy of 1.5 eV exceeded $3 \times 10^5 \text{ cm}^{-1}$. In contrast, the most pronounced changes in TES were observed in the heterostructures grown on semi-insulating substrates. In this case, the spectrum exhibited a distinct alteration in shape, characterized by two extrema of opposite polarity. This effect is attributed to the emergence of an additional internal electric field at the Bi/GaAs interface.

The most unexpected behavior was observed in the heterostructures based on Bi and p-GaAs. Only in this configuration, a significant increase in the emitted THz signals, relative to those from a bare p-GaAs substrate, was observed. This phenomenon is attributed to the hole drift velocity overshoot effect occurring in the strong internal electric field at the heterostructure. These findings highlight the potential of such structures for the development of efficient surface THz pulse emitters based on p-GaAs and other p-type semiconductors.

SUPPLEMENTARY MATERIAL

See the [supplementary material](#) that includes $\omega/2\theta$ XRD curves of structures consisting of 9 and 30 nm thick bismuth layers grown on semi-insulating GaAs confirming the presence of crystalline bismuth layers and revealed that its orientation corresponds to the 003 direction, assuming a hexagonal lattice. This behavior was also observed in other structures. Figure S2 shows THz pulses obtained from Bi/GaAs heterostructures with differing degrees of GaAs doping. The third figure illustrates the reflectance spectra of the investigated structures within a broad spectral range, measured in the same geometry as the THz excitation.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Vaidas Pačebutas: Investigation (equal); Methodology (equal); Resources (equal); Visualization (equal); Writing – review & editing (equal). **Ričardas Norkus:** Funding acquisition (equal); Investigation (equal); Visualization (equal); Writing – review & editing (equal). **Gintautas Tamulaitis:** Funding acquisition (equal); Writing – review & editing (equal). **Benas Stanionis:** Investigation (equal); Resources (equal); Writing – review & editing (equal). **Arūnas Krotkus:** Conceptualization (equal); Formal analysis (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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