Chapter 2
Optically Pumped Terahertz Radiation Sources Based on Impurity Carrier Transitions in Quantum Wells

Dmitry Firsov, Ivan Makhov, Vadim Panevin, Hayk A. Sarkisyan, and Leonid Vorobjev

Abstract A review of the results of studies of terahertz radiation associated with impurity electron transitions in n-doped $GaAs/AlGaAs$ quantum wells under conditions of interband optical excitation of nonequilibrium charge carriers is presented. The principles of radiation generation and methods of controlling its intensity are described: a decrease in the lifetime of electrons at impurity levels due to stimulated interband radiation and the introduction of a compensating acceptor impurity.

2.1 Introduction

Terahertz (THz) radiation (wavelength 30–300 μm) has a wide range of applications. High penetrating power for dry non-metallic objects allows its use in the field of non-destructive testing and in security systems. In the terahertz frequency range, many organic molecules have characteristic absorption bands, which makes it possible to create systems for the diagnosis of materials. Terahertz radiation is non-ionizing, unlike X-rays, which are used in medical diagnostics. It makes terahertz radiation safer for living organisms. Despite the vast areas of possible application of terahertz radiation, its use is significantly limited due to the difficulties with the creation of affordable compact and efficient sources of terahertz radiation.

To date, the most efficient and compact source of terahertz radiation is a quantum cascade laser, the operation principle of which is based on intersubband electron tran-

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D. Blaschke et al. (eds.), *Optics and Its Applications*, Springer Proceedings in Physics 281, https://doi.org/10.1007/978-3-031-11287-4_2

sitions during their vertical transport in nanostructures with tunnel-coupled quantum wells [1]. The most important factor limiting the widespread use of terahertz quantum cascade lasers is the extreme technological complexity of these devices. Therefore, the problem of finding and developing alternative mechanisms and schemes for generating terahertz radiation is urgent.

In n-type semiconductors, under conditions of optical interband excitation of nonequilibrium charge carriers, terahertz radiation can arise when nonequilibrium electrons are captured from the conduction band to donor impurity states, the depopulation of which occurs due to spontaneous electron-hole recombination during electron transitions from the ground donor state to the valence band. Terahertz radiation of this type was previously observed in bulk semiconductors [2–6]. Placing an impurity center in a quantum well opens up the possibility of controlling its energy spectrum due to the size quantization effect by changing the parameters of doped quantum wells and the possibility of controlling the frequency of impurity terahertz luminescence.

In this paper, we review the results of studying the terahertz impurity luminescence under the conditions of interband photoexcitation of nonequilibrium charge carriers in nanostructures with doped $GaAs/AlGaAs$ quantum wells. An increase in the intensity of the observed radiation can be obtained due to the effective depopulation of the ground donor state. To this end, two approaches were used: the fast depopulation of the ground impurity states by stimulated near-infrared (NIR) emission and a substantial compensation of impurities in quantum wells. In the latter case, the compensation of impurities leads to the appearance of an additional recombination channel of the "donor-acceptor" type, and also decreases the equilibrium population of impurity states.

2.2 Terahertz Photoluminescence Under Interband Photoexcitation of Quantum Wells Doped with Shallow Donors

2.2.1 Mechanism of Terahertz Emission

Let us consider the mechanism of impurity terahertz photoluminescence in quantum wells doped with shallow donors under interband optical excitation. A diagram illustrating optical transitions of charge carriers in doped quantum wells under interband photoexcitation is shown in Fig. 2.1. At a low temperature of the crystal lattice, the donor is neutral, i.e. in equilibrium, an electron is located in the ground state of the donor $1s$. Under interband optical pumping, depending on the photon energy of the exciting radiation, the generation of electron-hole pairs will occur either directly in the quantum wells or also in the barrier layers of the structure (this optical transition is marked with a solid arrow labeled "optical pumping" in Fig. 2.1). Then, nonequilibrium photoexcited electrons and holes, being thermalized, will descend to the lower subbands of the size quantization of electrons $e1$ and heavy holes $hh1$. After this,

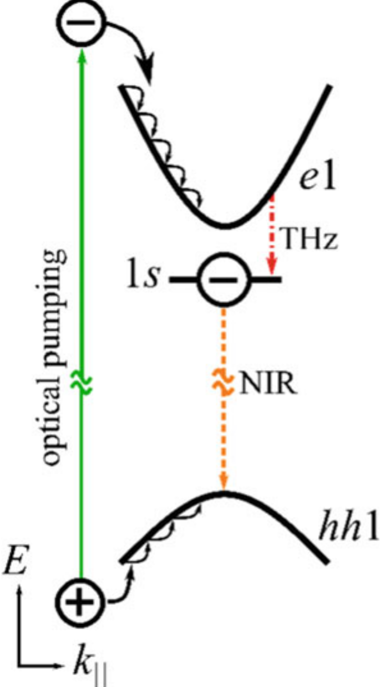

Fig. 2.1 Diagram of optical transitions of charge carriers in donor-doped quantum wells under interband optical excitation

the recombination of an electron in the ground state $1s$ of the donor impurity with a nonequilibrium hole from the $hh1$ subband is possible, accompanied usually by spontaneous emission of a photon corresponding to the near-infrared range. After the $1s$ donor ground state is depopulated, a nonequilibrium electron from the first electron subband $e1$ can be captured by an ionized donor with a spontaneous emission of a terahertz photon.

This section presents the results of the first studies of low-temperature impurity terahertz photoluminescence in nanostructures with $GaAs/AlGaAs$ quantum wells doped with shallow donors under conditions of interband optical excitation of nonequilibrium charge carriers.

2.2.2 Experimental Samples and Techniques

Investigations of impurity terahertz photoluminescence were performed on samples with quantum wells of various widths. Here, we present the results for a nanostructure containing 50 periods of $GaAs$ quantum wells of 30 nm, separated by tunnel-nontransparent $Al_{0.3}Ga_{0.7}As$ barriers with a thickness of 7 nm. Each quantum well of the nanostructure was doped with silicon in a narrow layer 4 nm wide, shifted from the center of the quantum well by 6 nm. The surface concentration of the dopant was

$3 \cdot 10^{10}$ cm^{-2}. The nanostructure had a 20 nm thick $GaAs$ cover layer doped with donors with a volume concentration of $5 \cdot 10^{17}$ cm^{-3}.

For measurements in the temperature range from 4 to 320 K, a Janis PTCM-4-7 optical closed-cycle cryostat was used. Interband photoexcitation of nonequilibrium charge carriers was carried out by modulated radiation of a continuous-wave solid-state Nd:YAG laser with frequency doubling in a lithium iodate crystal ($LiIO_3$) with a wavelength of 532 nm.

The terahertz photoluminescence emission of the sample was collected by an off-axis parabolic mirror of a Bruker Vertex 80v vacuum infrared Fourier spectrometer operating in the step-scan mode. In order to prevent the scattered pumping radiation from entering the measuring channel of the setup, a black polyethylene filter 100μm thick was installed in front of the entrance window of the Fourier spectrometer. An Infrared Laboratories, Inc., liquid helium-cooled silicon bolometer was used as a terahertz radiation detector. The photoresponse signal from the bolometer preamplifier was measured with a phase-sensitive detector lock-in amplifier SR-830 at the modulation frequency of the pumping radiation. The measured signal interferogram was converted into a spectrum using the OPUS software.

We also studied photoluminescence spectra in the near-infrared frequency range using an experimental setup based on a Horiba JobinYvon FHR 640 monochromator with a holographic grating consisted of 1200 groves per mm and a liquid nitrogen cooled silicon CCD matrix.

2.2.3 Experimental Results

When the structure is illuminated, optical generation of electron-hole pairs can occur not only in low-dimensional layers, but also in a semi-insulating GaAs substrate, which can contain residual impurities. In addition, nonequilibrium photoexcited charge carriers can reach the substrate due to diffusion from the active layers of the nanostructure, since the total thickness of the active layers of the nanostructure is comparable with the diffusion lengths of charge carriers in $GaAs$ [7]. As a result, two bands were found in the terahertz photoluminescence spectrum of the nanostructure, one of which is associated with impurities in the quantum wells. The second band, which was also observed in the sample, which is a substrate without epitaxial layers, is associated with optical transitions of charge carriers in the semi-insulating substrate of the structure. Note that the first band of terahertz photoluminescence cannot be caused by intersubband optical transitions of charge carriers in quantum wells. This is due to the fact that, in accordance with the selection rules, such optical transitions are allowed only for light polarized along the growth axis of the structure, while in the geometry of the experiment performed, radiation polarized in the plane of quantum wells was recorded.

The terahertz photoluminescence spectra of the nanostructure with quantum wells for various lattice temperatures are shown in Fig. 2.2 [11, 12].

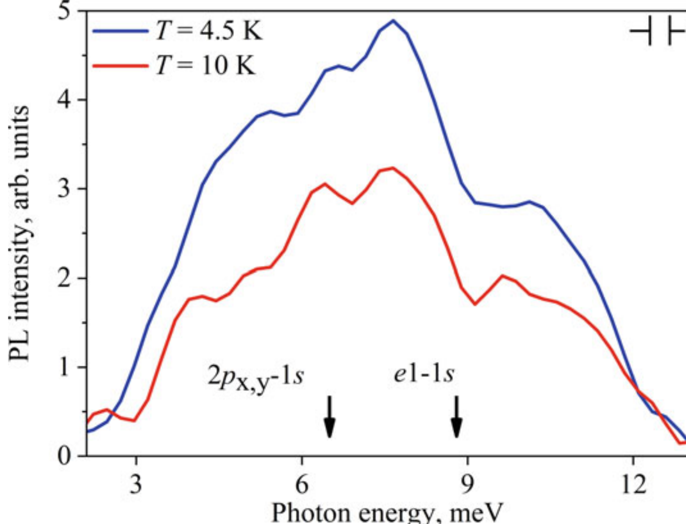

Fig. 2.2 Terahertz photoluminescence spectra of 30 nm $GaAs/AlGaAs$ quantum wells at various temperatures of the crystal lattice

In accordance with the calculation of the energy spectrum of shallow donors in $GaAs/Al_{0.3}Ga_{0.7}As$ QWs 30 nm wide, the binding energy of the donor impurity is about 9 meV [8]. Since the spectral position of the terahertz photoluminescence band of the nanostructure is close to the calculated value of the donor binding energy in quantum wells, the observed photoluminescence can be associated with optical transitions of nonequilibrium electrons with the participation of shallow donor states.

The observed radiation can be associated with several optical transitions of electrons. Firstly, this band can be caused by intracenter optical transitions of electrons between the excited $2p_{x,y}$ and ground $1s$ donor states. The calculated value of the transition energy is about 6.6 meV and is marked with an arrow $2p_{x,y}$-$1s$ in Fig. 2.2. Secondly, this terahertz photoluminescence band can be associated with optical transitions of electrons from the first electron subband $e1$ to the $1s$ ground state of the donor impurity. The calculated value of the optical transition energy $e1 - 1s$ is 8.8 meV and is indicated by the arrow $e1 - 1s$ in Fig. 2.2. The significant width of the spectrum is caused by the broadening of impurity states due to the high concentration of donor impurity centers [9], as well as by the broadening of the donor binding energy due to the finite width of the doping region of quantum wells. Narrow lines of impurity terahertz luminescence are observed in semiconductors with an impurity concentration much lower than in our case [10].

An increase in the lattice temperature from 4.5 K to 10 K leads to a 1.5-fold decrease in the integral luminescence intensity. Such quenching of impurity terahertz photoluminescence with increasing temperature has already been observed for the case of doped bulk semiconductors [3] and was attributed to a decrease in

the probability of capture of nonequilibrium charge carriers by ionized donor centers with increasing lattice temperature. In addition, the decrease in the intensity of impurity terahertz photoluminescence with an increase in the lattice temperature can be caused by the expansion of the distribution function of charge carriers in the QW subband with temperature.

The photoluminescence spectra of the near-infrared range were also investigated. In accordance with the mechanism of the observed terahertz impurity luminescence, these spectra exhibit features associated with radiative electron-hole recombination between the ground donor state and the first hole subband of the quantum well.

Similar measurements were performed for a nanostructure with narrower quantum wells 16.1 nm wide [13]. The calculated value of the binding energy of the donor impurity of silicon in such quantum wells is about 10 meV, which led to a short-wavelength shift of the impurity terahertz photoluminescence band relative to the impurity luminescence band of wide quantum wells and to the corresponding modification of the photoluminescence spectra of the near-infrared range.

2.3 Influence of Stimulated Near-Infrared Radiation on Terahertz Photoluminescence

2.3.1 Introduction

According to the proposed mechanism of terahertz impurity photoluminescence, the depopulation of the donor ground state occurs due to spontaneous radiative or nonradiative recombination during transitions of charge carriers between the donor ground state and the states of the first hole subband. There are a number of approaches that make it possible to increase the efficiency of impurity terahertz photoluminescence in doped quantum wells. One of them is an increase in the rate of depletion of the final state for the impurity terahertz transition of an electron (i.e., the ground donor state), since the capture of nonequilibrium electrons to the ground impurity state, accompanied by the emission of photon in the terahertz range, in particular, is determined by the population of the ground donor state. An increase in the rate of depopulation of the ground donor state can be achieved due to the organization of stimulated emission in the near-infrared range in the same nanostructure, namely, the organization of stimulated optical transitions of charge carriers from the ground state of the donor to the hole subband.

A similar mechanism has already been demonstrated earlier in [14, 15] when observing mid-infrared radiation from laser diode structures with vertically coupled self-organized $InGaAs/AlGaAs$ quantum dots and $InGaAs/GaAs$ quantum wells. Laser generation, realized in the near-infrared range of the spectrum in structures with quantum dots during transitions of charge carriers between the lower electron and hole levels, led to an increase in the intensity of spontaneous intraband transitions of charge carriers between the levels of quantum dots, accompanied by

the emission of photons in the mid-infrared range. At the same time, in the absence of lasing in the near-infrared range of the spectrum, spontaneous emission of the mid-infrared region was not observed at all in structures with quantum dots. In structures with quantum wells, spontaneous emission of mid-infrared radiation associated with intersubband transitions of nonequilibrium charge carriers in the wells was also observed, but it did not have a threshold character as in structures with quantum dots.

This section presents the results of studies of the spectra of spontaneous terahertz photoluminescence in laser nanostructures with doped quantum wells with a waveguide for near-infrared radiation. When lasing in the near-infrared range occurs in such a nanostructure, the rate of depopulation of the ground donor state increases due to stimulated transitions from the ground donor state to the first hole subband, which in turn should be reflected in an increase in the intensity of terahertz radiation when electrons are captured from the first electron subband and excited donor states to the ground donor state.

2.3.2 Experimental Samples and Techniques

We studied a nanostructure with doped quantum wells with waveguide layers, as well as a semi-insulating $GaAs$ substrate, which did not contain epitaxial layers. A sample of a nanostructure with quantum wells was grown by molecular beam epitaxy on a semi-insulating $GaAs$ substrate. The quantum wells were formed by 7.6 nm thick $GaAs$ layers separated by 5 nm thick $Al_{0.3}Ga_{0.7}As$ barriers. Doping of quantum wells with a surface concentration of $5 \cdot 10^{10}$ cm^{-2} was carried out with silicon in the central region of each quantum well with a width of 2.6 nm. In total, the nanostructure contained 10 quantum wells, which were placed in a symmetric gradient waveguide for near-infrared radiation, formed by wide-gap gradient $Al_xGa_{1-x}As$ layers (composition x varied from 0.4 to 0.6) 0.6 μm thick. The nanostructure had a 10 nm thick $GaAs$ cover layer doped with silicon with a volume concentration of $5 \cdot 10^{17}$ cm^{-3}.

To get lasing in the near-infrared range, the $GaAs$ semi-insulating substrate was grinded down to a thickness of about 100 μm, and then high-Q cavity of total internal reflection with sides of about $600 \cdot 600$ μm^2 was cleaved out of it.

During the experiment, the samples were placed on a copper holder in a Janis PTCM-4-7 closed cycle optical cryostat, which made it possible to change the sample temperature in the range of $4 - 320$ K. The photoluminescence studies of both terahertz and near-infrared ranges were carried out simultaneously. The pump radiation was directed to the sample in a cryostat using a system of mirrors and a spherical lens. In this case, the lens was installed in a defocused position to illuminate the entire surface of the laser nanostructure resonator. Terahertz photoluminescence radiation was collected by an off-axis parabolic mirror of a Fourier spectrometer from the sample surface. Spontaneous and stimulated near-infrared radiation was collected from the facet of the nanostructure cavity using a system of lenses and mirrors and directed to the entrance slit of the grating monochromator.

To achieve lasing in the near-infrared spectral range, high-power optical excitation of the samples was carried out by a pulsed solid-state Nd:YAG laser with frequency doubling in a $LiIO_3$ crystal. The pumping radiation parameters are as follows: the radiation wavelength is 532 nm, the pulse duration is 250 ns, and the repetition rate is 8 kHz. The intensity of the polarized pumping laser radiation was varied using an adjustable attenuator consisting of a combination of a half-wave plate and a Glan-Taylor prism, and was additionally modulated by a mechanical chopper at a frequency of 87 Hz. The need for additional modulation of the pulsed pump radiation at a low frequency is caused by the limited bandwidth of the bolometer, which acts as a photodetector of terahertz radiation. The photoresponse signal of bolometer was measured with a phase-sensitive lock-in amplifier SR-830 at a frequency of 87 Hz.

Except for the details described above, the technique for studying the terahertz and near-infrared photoluminescence spectra of samples does not differ from the technique described in Sect. 2.2.2.

2.3.3 Experimental Results, Their Analysis and Discussion

The spectral dependence of the terahertz photoluminescence intensity measured at a low pulsed optical excitation power of 40 W/cm^2 for a nanostructure with doped quantum wells and a waveguide for near-infrared radiation, prepared in the form of a total internal reflection cavity, is shown in Fig. 2.3 [21]. In this case, in the spectra of spontaneous near-infrared photoluminescence, an emission band is observed, associated with radiative electron-hole recombination through donor states. As can be seen, in the terahertz photoluminescence spectrum, one emission band is observed with a maximum intensity near the photon energy of 20 meV. Note, that, under the same conditions, terahertz radiation from a semi-insulating $GaAs$ substrate was not observed.

In accordance with the calculation of the energy spectrum of donor states [16], the binding energy of a donor impurity located in the center of a $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well with a width of 7.6 nm is approximately 13.5 meV. Therefore, the observed band of terahertz radiation with a maximum intensity near the photon energy of 20 meV (see Fig. 2.3) is associated with optical transitions of nonequilibrium electrons from the first electron subband $e1$ to the ground state of the donor impurity $1s$.

A significant shift of the spectral position of the intensity maximum of the terahertz photoluminescence band from the binding energy of the donor impurity in the quantum well can be explained as follows. It is known that the wave functions of electrons on donor impurity states are formed mainly by the wave functions of the conduction band. Therefore, optical transitions of electrons between the $1s$ donor ground state and the first electron subband e1 are forbidden in the standard dipole approximation, since the wave functions of the initial and final states in this case have the same symmetry. However, this condition is strictly fulfilled only for the zero value of the wave vector of the electron. Theoretical calculations of the spec-

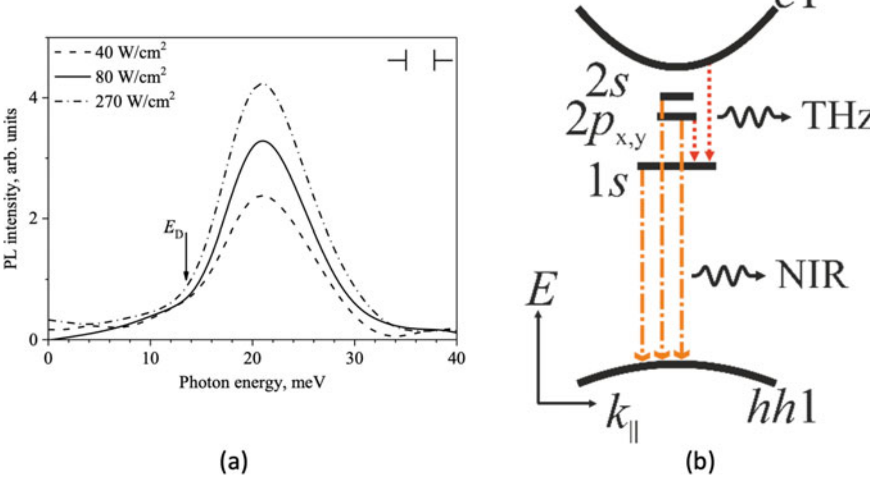

Fig. 2.3 Terahertz photoluminescence spectra of a nanostructure with doped quantum wells, measured at a temperature $T = 5$ K and various optical excitation powers (**a**); Diagram of impurity optical transitions of charge carriers in quantum wells. Dotted and dash-dotted arrows show optical transitions in the terahertz and near-infrared ranges of the spectrum, respectively (**b**)

tral dependence of the photoionization cross section (which is proportional to the photodeionization cross section [17]) of shallow donors in quantum wells performed in [18] showed that the maximum value of the donor photoionization cross section in quantum wells is located at photon energies exceeding the binding energy of the donor impurity. Thus, the maximum of the photodeionization cross section, which determines the shape of the spectrum of impurity photoluminescence associated with the $e1$-$1s$ transitions, should be shifted to the short-wavelength region of the spectrum relative to the binding energy of the donor impurity, which agrees with the observed spectrum of terahertz photoluminescence (see Fig. 2.3a).

The next step was to increase the optical excitation power of the nanostructure, which should lead to the appearance of stimulated near-infrared radiation during donor impurity transitions of charge carriers. This, in turn, should lead to a more efficient depopulation of the ground donor state, which is the final state for terahertz electron transitions. Figure 2.4 shows the dependence of the intensity of the near-infrared radiation, integrated over the entire photoluminescence spectrum, on the optical excitation power. This dependence has a threshold character, which indicates the appearance of lasing in the near-infrared range of the spectrum. The threshold value of the pumping power for the appearance of lasing is about 120 W/cm^2.

In the near-infrared photoluminescence spectra, when the pumping power reaches 120 W/cm^2, a high-intensity narrow emission line appears against the background of a broad photoluminescence spectrum. Based on the results of calculating the energy spectrum of donor states in quantum wells, the arising stimulated emission line is

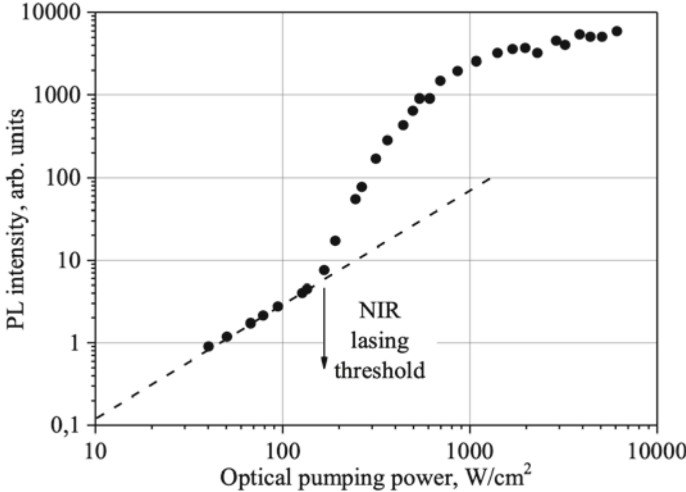

Fig. 2.4 Dependence of the integrated intensity of the near-infrared photoluminescence of a nanostructure with doped quantum wells on the optical excitation power

associated with the radiative electron-hole recombination between the excited state of the donor $2s$ and the first hole subband $hh1$ (see Fig. 2.3 b).

Lasing in the near-infrared range initially occurs during optical transitions of charge carriers with the participation of an excited rather than a ground donor state. This may be due to the different gain for optical transitions with the participation of the excited and ground donor states.

The dependence of the integrated intensity of terahertz radiation on the power of optical excitation of the nanostructure is shown in Fig. 2.5.

It can be seen from this dependence that at low excitation levels, i.e. at pumping powers not exceeding the near-infrared lasing threshold (the threshold value is indicated by the arrow $2s$-$hh1$ in Fig. 2.5), the integral intensity of the impurity terahertz photoluminescence is proportional to the square root of the optical pumping power (the square root dependence is shown by the dashed line in Fig. 2.5). As follows from the terahertz photoluminescence spectra measured at low pumping powers (see Fig. 2.3a), an increase in the terahertz photoluminescence intensity in this range of photoexcitation powers is associated with an increase in the intensity of the $e1$-$1s$ band. This dependence of the terahertz impurity photoluminescence intensity on the pump power is in a good agreement with the theoretical and experimental results for terahertz impurity radiation upon interband optical pumping of bulk semiconductors [4].

When the pumping power corresponding to the onset of near-infrared lasing at $2s - hh1$ transitions is reached, the rate of increase in the terahertz photoluminescence intensity associated with optical transitions $e1$-$1s$ begins to decrease (see Fig. 2.5). The intensity of such terahertz photoluminescence I^{e1}_{THz}, associated with

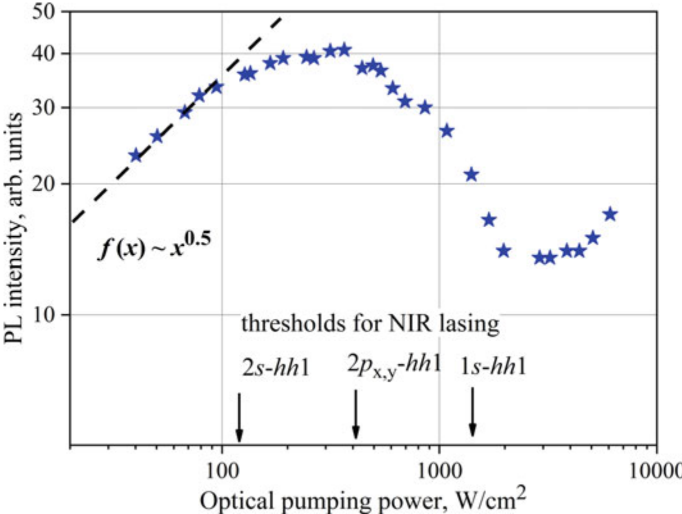

Fig. 2.5 Dependence of the integrated intensity of terahertz photoluminescence of a nanostructure with doped quantum wells on the level of interband optical excitation, measured at a temperature of $T = 5$ K

electron transitions $e1$-$1s$, in the first approximation depends on the concentration of electrons $n\,(e1)$ in the $n^+\,(1s)$ subband and the concentration of free donor states $1s$:

$$I^{e1}_{THz} \propto n\,(e1) \cdot n^+\,(1s)\,. \tag{2.1}$$

Without taking into account the compensation of impurities, the concentration $n^+(1s)$ of free donor $1s$ states can be determined as follows:

$$n^+\,(1s) = N_D - n\,(1s) - n\left(2p_{x,y}\right) - n\,(2s)\,, \tag{2.2}$$

where N_D is the total donor concentration in quantum wells, and $n\,(1s)$, $n\left(2p_{x,y}\right)$, and $n\,(2s)$ are the quasi-equilibrium electron concentrations in donor states $1s$, $2p_{x,y}$ and $2s$, respectively. The presence of the last two terms in expression (2.1) is caused by the fact that the considered donor impurity centers are singly charged, i.e. one donor impurity center cannot capture more than one electron [19]. The concentrations $n\,(e1)$ and $n^+\,(1s)$ themselves are determined by the rates of capture of electrons from the first electron subband $e1$ to excited and ground donor states and the rates of recombination of electrons from donor states with holes from the valence subbands. Probably, the arising stimulated emission line caused by stimulated impurity transitions $2s$-$hh1$ leads to a faster depopulation of the excited donor state $2s$ in the QW compared to depletion due to spontaneous transitions, which leads to the corresponding more efficient nonradiative capture of electrons from the first electron subband $e1$ into an excited donor state $2s$. It follows that, with the onset of stimulated $2s$-$hh1$ transitions, the rate of increase in the electron concentration in the subband

n ($e1$) with increasing pumping power can decrease, as well as a change in the concentration n^+ ($1s$) of free donor states $1s$ capable of capturing an electron from the $e1$ subband.

A subsequent increase in the optical excitation power to a level of 400 W/cm^2 leads to a decrease in the integrated intensity of the impurity terahertz photoluminescence with an increase in the level of optical excitation of the nanostructure (see Fig. 2.5). As follows from the corresponding terahertz photoluminescence spectra measured at photoexcitation powers exceeding 270 W/cm^2, the decrease in the integrated terahertz photoluminescence intensity is accompanied by a decrease in the intensity of the terahertz luminescence band associated with optical transitions of electrons $e1$-$1s$.

The decrease in the integrated intensity of impurity terahertz photoluminescence at pumping powers above 400 W/cm^2 is accompanied by the appearance of an additional line of impurity stimulated near-infrared emission, observed in the corresponding spectra. The appearance of the second line of impurity stimulated emission is associated with stimulated transitions of charge carriers $2p_{x,y}$-$hh1$. Such transitions probably lead to a faster nonradiative capture of nonequilibrium electrons from the first electron subband $e1$ to excited donor states $2p_{x,y}$, which in turn leads to a decrease in the intensity of impurity terahertz electron transitions $e1$-$1s$. This is due to a change in the concentration of electrons in the first electron subband and free ground donor states included in expression (2.1), by analogy with the effect of stimulated transitions $2s$-$hh1$ on the intensity of terahertz photoluminescence at transitions $e1$-$1s$.

A further increase in the optical excitation power above 1 kW/cm^2 leads to the appearance of a third line of stimulated near-infrared radiation, which is associated with radiative recombination of electrons from the ground donor state with nonequilibrium holes from the first subband of heavy holes $hh1$.

It is the onset of stimulated optical transitions $1s$-$hh1$ that should lead to the effective depopulation of the $1s$ donor ground state and, accordingly, to an increase in the intensity of impurity terahertz photoluminescence. Indeed, the dependence of the integral intensity of the impurity terahertz photoluminescence on the optical excitation power shows an increase in the terahertz photoluminescence intensity with increasing pumping at photoexcitation powers exceeding 2 kW/cm^2 (see Fig. 2.5), i.e. exceeding the threshold for lasing at impurity transitions $1s$-$hh1$.

It is noteworthy that in the terahertz photoluminescence spectra, the appearance of stimulated emission in the near-infrared range during the $1s$-$hh1$ transitions leads to the appearance of a new terahertz radiation line near the photon energy of 9 meV. In accordance with the results of calculating the energy spectrum of donor impurity states in our quantum wells [16], a new terahertz radiation line at a photon energy of 9 meV is associated with intracenter optical transitions of nonequilibrium electrons between the excited $2p_{x,y}$ and ground $1s$ donor states in quantum wells. Probably, at pump powers exceeding 2 kW/cm^2, the ground donor states $1s$ begin to depopulate by near-infrared stimulated radiation faster than excited states $2p_{x,y}$, which leads to the appearance of radiative transitions $2p_{x,y}$-$1s$, accompanied by the emission of terahertz photons.

2.4 Terahertz Photoluminescence in Compensated Quantum Wells

2.4.1 Introduction

There is another way to increase the intensity of terahertz photoluminescence associated with impurity transitions of electrons in quantum wells. The intensity of the impurity terahertz photoluminescence depends on the population of the donor ground state, which is final for terahertz electron transitions. In this section, we consider the possibility of increasing the intensity of impurity terahertz radiation due to additional doping of quantum wells with acceptors, i.e., due to compensation of donors with acceptors. Firstly, such compensation of donor impurities by acceptor impurities will significantly lower the equilibrium population of donor states. Secondly, an additional recombination channel will arise for electrons from the $1s$ ground donor state due to recombination of the donor-acceptor type. The above factors should increase the intensity of impurity terahertz photoluminescence for compensated quantum wells as compared to uncompensated ones. In compensated quantum wells, emission bands associated with donor-acceptor recombination and recombination of the "first electron subband-acceptor" type can appear in the photoluminescence spectra of the near-infrared range. In the terahertz photoluminescence spectra, an emission band associated with the capture of holes from the first hole subband into acceptor states can also be detected.

2.4.2 Experimental Samples and Techniques

On semi-insulating $GaAs$ substrates, two structures were grown, each containing 50 periods of quantum wells formed by $GaAs$ layers 7.6 nm wide and separated by 5 nm wide $Al_{0.3}Ga_{0.7}As$ barriers. Quantum wells in the first nanostructure were doped only with a donor impurity (Si) with a surface concentration of $3 \cdot 10^{10}$ cm^{-2}. The quantum wells in the second nanostructure, in addition to the same donor impurity, contained a compensating acceptor (Be) impurity with the same concentration. Doping was carried out into the central region of the quantum wells 2.6 nm wide. Both nanostructures had a 5 nm thick $GaAs$ cover layer doped with silicon with a volume concentration of $5 \cdot 10^{17}$ cm^{-3}.

The technique for studying the photoluminescence of the near-infrared and terahertz spectral ranges in the described nanostructures is completely analogous to the technique described in Sect. 2.2.2.

In order to compare the terahertz photoluminescence intensities of two nanostructures, the dependences of the integrated terahertz photoluminescence intensity on the optical excitation power were measured. For these studies, a Ge:Ga photoresistor sensitive to terahertz radiation (sensitivity range $8 - 30$ meV) was installed opposite to the surface of the sample placed on a copper holder in a closed-cycle

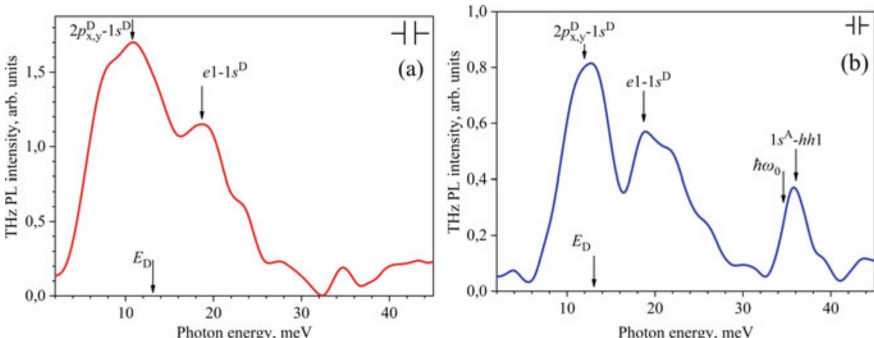

Fig. 2.6 Terahertz photoluminescence spectra of nanostructures with quantum wells doped with donors (**a**) and donors and acceptors (**b**), measured at a temperature of $T = 8$ K. The scales of the vertical axes in figures (**a**) and (**b**) differ

cryostat. The distance between the sample and the $Ge{:}Ga$ photodetector was about 12 mm. Thus, upon optical pumping of the sample, the integrated intensity of terahertz radiation from the sample surface was recorded by a $Ge{:}Ga$ photodetector. In order to prevent penetration of the reflected and scattered pumping radiation, as well as the near-infrared radiation of the photoluminescence of the sample, to the $Ge{:}Ga$ photodetector, cold filters made of black polyethylene (approximately 100 μm thick) and high-resistive germanium compensated with antimony and gold (1 mm thick) were installed at the input of the photodetector.

2.4.3 Experimental Results, Their Analysis and Discussion

In the spectra of near-infrared photoluminescence for both nanostructures with doped quantum wells, an emission band appears, associated with optical transitions of charge carriers through donor impurity states [25]. In addition, for a nanostructure with compensated quantum wells, the spectrum contains emission bands associated with impurity optical transitions of charge carriers between the first electron subband and acceptor states, as well as between the states of donors and acceptors in quantum wells.

The terahertz photoluminescence spectrum of a nanostructure with $GaAs/AlGaAs$ quantum wells doped only by donors is shown in Fig. 2.6a.

In the photoluminescence spectrum, two emission bands are observed with intensity maxima near the photon energies of 10 and 21 meV. No terahertz photoluminescence signal from the semi-insulating $GaAs$ substrate was observed under similar experimental conditions; therefore, the observed emission bands are caused by optical transitions of nonequilibrium charge carriers in low-dimensional doped layers. The detected bands of terahertz radiation cannot also be caused by intersubband transitions of charge carriers in quantum wells, since they are forbidden for radiation

polarized in the plane of quantum wells (it is this polarization of radiation that was used in experiments).

In accordance with the results of calculating the energy spectrum of the states of shallow donors in quantum wells $GaAs/Al_{0.3}Ga_{0.7}As$ [16], the binding energy of the ground donor state in quantum wells 7.6 nm wide is about 13.5 meV (this value is indicated by the arrow E_D in Fig. 2.6 a). Thus, the terahertz photoluminescence band with a maximum intensity near the quantum energy of 21 meV can be associated with optical transitions of nonequilibrium electrons from the first electronic subband $e1$ to the ground donor state $1s$. The corresponding luminescence band is indicated by the arrow $e1$-$1s^D$ in Fig. 2.6a.

The second band of terahertz photoluminescence with a maximum intensity near a quantum energy of 10 meV is probably associated with intracenter optical transitions of electrons between the excited $2p_{x,y}$ and ground $1s$ states of donors in quantum wells. This luminescence band is marked with an arrow $2p^D_{x,y}$-$1s^D$ in Fig. 2.6a. The spectral position of the observed band caused by intracenter transitions of electrons is in a good agreement with the results of calculating the energy spectrum of shallow donors in quantum wells from [16], according to which the energy gap between the ground $1s$ and excited $2p_{x,y}$ donor states is about 10 meV.

In the terahertz photoluminescence spectra of a nanostructure with quantum wells doped with both donors and acceptors, emission bands associated with optical transitions of charge carriers with the participation of both donor and acceptor impurity states should appear. Such a spectrum is shown in Fig. 2.6 b on the same line for the crystal lattice temperature of the nanostructure $T = 8$ K. Since, in the $6 - 28$ meV photon energy range, the emission spectrum of this nanostructure exhibits terahertz luminescence bands similar to those found for a nanostructure with quantum wells doped only by donors (see Fig. 2.6a), then emission in this spectral range is also associated with optical transitions of nonequilibrium photoexcited electrons from the first electron subband $e1$ and excited donor states $2p_{x,y}$ to the ground donor state $1s$. These bands of impurity terahertz photoluminescence are marked with arrows $2p^D_{x,y}$-$1s^D$ and $e1$-$1s^D$ in Fig. 2.6b.

The terahertz photoluminescence band, located near the photon energy of 36 meV, is observed only for a nanostructure with quantum wells, in which donors are compensated by acceptors, which makes it possible to associate this terahertz radiation band with the presence of acceptor impurities in the quantum wells. The binding energy of a beryllium acceptor located in the center of a $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well 7.6 nm wide is about 35 meV [22]. Therefore, the terahertz photoluminescence band near a photon energy of 36 meV is associated with optical transitions of nonequilibrium electrons between the ground acceptor state and the first subband of size quantization of heavy holes $hh1$. This photoluminescence band is marked with an arrow $1s^A$-$hh1$ in Fig. 2.6b on the same line. It should be noted that the true shape of the terahertz photoluminescence band associated with the presence of acceptor impurity states in quantum wells may differ from that presented. This is caused by the possible influence of lattice absorption in the $GaAs$ layers, since the energy of the transverse optical phonon in $GaAs$ is about 33.8 meV [23], which is very close

to the binding energy of the beryllium impurity in quantum wells. The energy of a transverse optical phonon is marked with an arrow $\hbar\omega_0$ in Fig. 2.6b.

Next, we compared the intensities of terahertz photoluminescence associated with donor impurity transitions of nonequilibrium electrons in quantum wells of both types. Comparative studies of the integrated intensity of terahertz photoluminescence for two nanostructures were performed using $Ge{:}Ga$ photoresistor, which is sensitive to terahertz radiation in the 8–30 meV photon energy range [24] and does not detect short-wavelength radiation associated with optical transitions with participation of acceptors.

The dependences of the integrated terahertz photoluminescence intensity on the optical pumping power in both studied nanostructures, measured in a wide range of photoexcitation powers at a grating temperature $T = 8$ K look similar. However, a nanostructure in which donors in quantum wells are compensated by acceptors demonstrates an order of magnitude greater integrated intensity of terahertz photoluminescence associated with donor impurity electron transitions, compared to a nanostructure in which shallow donors in quantum wells are not compensated. This is observed in the sufficiently wide range of photoexcitation powers.

2.5 Conclusion

This paper presents the results of experimental studies of low-temperature terahertz photoluminescence associated with impurity transitions of nonequilibrium charge carriers in quantum wells under interband optical excitation.

The spectral dependences of the intensity of spontaneous terahertz photoluminescence under interband optical pumping of nanostructures with donor-doped $GaAs/AlGaAs$ quantum wells of various widths are studied for the first time. Two approaches have been implemented to increase the efficiency of terahertz impurity luminescence:

- Increase in the rate of depletion of the ground donor state due to stimulated radiation in the near infrared range, implemented in the same nanostructure.
- Compensation of donor with acceptors, which leads to a decrease in the equilibrium population of donor states and to an additional depopulation of the donor ground state due to electron transitions from the donor ground state to acceptor states.

Acknowledgements Financial support from the Ministry of Science and Higher Education of the Russian Federation (state assignment) is gratefully acknowledged. IM also acknowledges a support from the Basic Research Program of the National Research University Higher School of Economics.

References

1. Köhler, R., Tredicucci, A., Beltram, F., Beere, H.E., Linfield, E.H., Davies, A.G., Ritchie, D.A., Iotti, R.C., Rossi, F.: Terahertz semiconductor-heterostructure laser. Nature **417**, 156–159 (2002)
2. Andrianov, A.V., Zakhar'in, A.O., Ivanov, Y.L., Kipa, M.S.: Terahertz impurity luminescence under the interband photoexcitation of semiconductors. JETP Lett. **91**(2), 96–99 (2010)
3. Zakhar'in, A.O., Andrianov, A.V., Egorov, AYu., Zinov'ev, N.N.: Terahertz photoluminescence from GaAs doped with shallow donors at interband excitation. Appl. Phys. Lett. **96**(21), 211118 (2010)
4. Zakhar'in, A.O., Bobylev, A.V., Andrianov, A.V.: Terahertz emission upon the interband excitation of GaN layers. Semiconductors **46**(9), 1135–1139 (2012)
5. Andrianov, A.V., Zakhar'in, A.O., Zhukavin, R.K., Shastin, V.N., Abrosimov, N.V., Bobylev, A.V.: Terahertz intracenter photoluminescence of silicon with lithium at interband excitation. JETP Lett. **100**(12), 771–775 (2015)
6. Andrianov, A.V., Zakhar'in, A.O., Petrov, A.G.: Intraexciton and intracenter terahertz radiation from doped silicon under interband photoexcitation. JETP Lett. **107**(9), 540–543 (2018)
7. Casey, H.C., Miller, B.I., Pinkas, I.: Variation of minority-carrier diffusion length with carrier concentration in GaAs liquid-phase epitaxial layers. J. Appl. Phys. **44**(3), 1281–1287 (1973)
8. Firsov, D.A., Shalygin, V.A., Panevin, V.Yu., Melentyev, G.A., Sofronov, A.N., Vorobjev, L.E., Andrianov, A.V., Zakhar'in, A.O., Mikhrin, V.S., Vasil'ev, A.P., Zhukov, A.E., Gavrilenko, L.V., Gavrilenko, V.I., Antonov, A.V., Aleshkin, V.Ya.: Terahertz emission and photoconductivity in n-type GaAs/AlGaAs quantum wells: the role of resonant impurity states. Semiconductors **44**(11), 1394–1397 (2010)
9. Kogan, Sh.M., Lifshits, T.M.: Photoelectric spectroscopy—a new method of analysis of impurities in semiconductors. Phys. Status Solidi A **39**(1), 11–39 (1977)
10. Melngailis, I., Stillman, G.E., Dimmock, J.O., Wolfe, C.M.: Far-infrared recombination radiation from impact-ionized shallow donors in GaAs. Phys. Rev. Lett. **23**(19), 1111–1114 (1969)
11. Vorobjev, L.E., Firsov, D.A., Panevin, V.Y., Sofronov, A.N., Balagula, R.M., Makhov, I.S.: Near- and far-infrared emission from GaAs/AlGaAs quantum wells under interband optical excitation. St. Petersburg State Polytechnical Journal. Phys. Math. **4**(182), 109–114 (2013)
12. Firsov, D.A., Vorobjev, L.E., Panevin, V.Y., Sofronov, A.N., Balagula, R.M., Makhov, I.S., Kozlov, D.V., Vasil'ev, A.P.: Terahertz radiation associated with the impurity electron transition in quantum wells upon optical and electrical pumping. Semiconductors **49**(1), 28–32 (2015)
13. Vinnichenko, M.Ya., Makhov, I.S., Panevin, V.Yu., Sofronov, A.N., Firsov, D.A., Vorobjev, L.E., Sadofev, Y.G., Vasiliev, A.P.: Terahertz radiation related to the electron relaxation after interband optical pumping in doped quantum wells. In: International Conference on Infrared, Millimeter, and Terahertz Waves 2016, 7758782 (2016)
14. Vorob'ev, L.E., Firsov, D.A., Shalygin, V.A., Tulupenko, V.N., Shernyakov, Yu.M., Ledentsov, N.N., Ustinov, V.M., Alferov, Zh.I.: Spontaneous far-IR emission accompanying transitions of charge carriers between levels of quantum dots. JETP Lett. **67**(4), 275–279 (1998)
15. Vorob'ev, L.E., Firsov, D.A., Shalygin, V.A., Tulupenko, V.N., Ledentsov, N.N., Kop'ev, P.S., Ustinov, V.M., Shernyakov, Yu.M., Alferov, Zh.I.: The outlook for the development of radiation sources in the middle-IR range based on the intraband transitions between the energy levels of charge carriers in injection laser heterostructures with quantum dots and wells. Physics-Uspekhi **42**(4), 391–405 (1999)
16. Mailhiot, C., Chang, Y.-C., McGill, T.C.: Energy spectra of donors in $GaAs - Ga_{1-x}Al_xAs$ quantum well structures in the effective-mass approximation. Phys. Rev. B **26**(8), 4449–4457 (1982)
17. Ridley, B.K.: Quantum processes in semiconductors, 5th edn. Oxford University Press, New York (2013)
18. El-Said, M., Tomak, M.: Photoionization of impurities in infinite-barrier quantum wells. J. Phys. Chem. Solids **52**(4), 603–606 (1991)

19. Blakemore, J.S.: Semiconductor statistics. Pergamon Press, Oxford (1962)
20. Makhov, I.S., Panevin, V.Yu., Sofronov, A.N., Firsov, D.A., Vorobjev, L.E., Vinnichenko, M.Ya., Vasil'ev, A.P., Maleev, N.A.: The effect of stimulated interband emission on the impurity-assisted far-infrared photoluminescence in GaAs/AlGaAs quantum wells. Superlattices Microstruct. **112**, 79–85 (2017)
21. Makhov, I.S., Panevin, VYu., Firsov, D.A., Vorobjev, L.E., Vasil'ev, A.P., Maleev, N.A.: Terahertz photoluminescence of the donor doped GaAs/AlGaAs quantum wells controlled by the near-infrared stimulated emission. J. Lumin. **210**, 352–357 (2019)
22. Zheng, W.M., Halsall, M.P., Harmer, P., Harrison, P., Steer, M.J.: Acceptor binding energy in δ-doped GaAs/AlAs multiple-quantum wells. J. Appl. Phys. **92**(10), 6039–6042 (2002)
23. Blakemore, J.S.: Semiconducting and other major properties of gallium arsenide. J. Appl. Phys. **53**(10), R123–R181 (1982)
24. Haller, E.E., Hueschen, P.L., Richards, P.L.: Ge: Ga photoconductors in low infrared backgrounds. Appl. Phys. Lett. **34**(8), 495–497 (1979)
25. Makhov, I.S., Panevin, VYu., Firsov, D.A., Vorobjev, L.E., Klimko, G.V.: Impurity-assisted terahertz photoluminescence in compensated quantum wells. Appl. Phys. Lett. **126**(19), 175702 (2019)