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Terahertz photoluminescence from GaAs doped with shallow donors at interband excitation

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We report on the observation of efficient generation of terahertz radiation at continuous-wave interband excitation of n-GaAs at low temperatures. The radiative transitions, accompanying relaxation and trapping of photoexcited electrons to localized donor states or to empty states in impurity subband, lead to the emission of terahertz photons with a relatively high external quantum yield up to 0.3%. © *2010 American Institute of Physics*. [doi:10.1063/1.3441401]

The growth of interest to the studies of optical phenomena leading to generation of electromagnetic terahertz (THz) radiation is stipulated by a vast number of applications in physics, chemistry, biophysics, and live sciences awaiting compact and efficient THz sources. One of the schemes to generate THz radiation relies upon optical transitions between the energy levels of shallow impurities in semiconductors and has been demonstrated under the conditions of impact ionization of impurity centers[1–6] or impurity photoionization by optical pumping the transition "impurity ground state-conduction or valence band."[7]

Here we introduce, different than cited above, mechanism of THz emission exploiting band-to-band excitation of GaAs doped with shallow donors. This mechanism is closely related to the recombination of nonequilibrium holes with the electrons of neutral donors (Fig. 1). Such a recombination process produces an intensive photoluminescence (PL) line D^0h observed in the spectra of the near band edge emission of n-GaAs crystals at low temperatures (see, for example, Ref. 8 and other references therein). The remaining nonequilibrium electrons in conduction band are bound to charged donors that were depopulated with the $D^0 \rightarrow h$ recombination process. The process is completed by cascading trapping of the nonequilibrium electrons from the continuum states of the conduction band to the bound discrete hydrogenlike states accompanied by emission of acoustic phonons (nonradiative channel) and THz photons (radaitive channel). It is this radiative process that gives rise to THz PL. In the present paper, we report on the experimental observation of the THz emission under band-to-band photoexcitation of n-GaAs at helium temperatures, present results of spectroscopic study of the emission and discuss the optical transitions responsible for the THz PL.

Basic experiments were carried out on two types of GaAs samples. The samples of the first type were GaAs layers of 10 μm thickness grown by liquid-phase epitaxy (LPE) on 300 μm thick GaAs semi-isolating substrate. These layers were not deliberately doped but had electron concentration of 5×10^{15} cm^{-3} at $T=300$ K ($N_D-N_A\sim5\times10^{15}$ cm^{-3}). The samples of the second type were MBE grown GaAs layers on semi-isolating substrate. These layers were doped with Si and the doping level corresponded to electron concentration of 8×10^{16} cm^{-3} at $T=300$ K ($N_D-N_A\sim8\times10^{16}$ cm^{-3}). The thickness of the n-GaAs layers was 1.5 μm, the substrate thickness was 300 μm. Since the critical impurity concentration N_D-N_A for the Mott transition for n-GaAs is $\sim1.6\times10^{16}$ cm^{-3},[9] the samples of the first type were below the Mott transition, at the nonmetallic range, while the samples of the second type were above that, pertaining to the metallic range. Samples under study were mounted in a He-cryostat with the temperature control modified for the optical spectroscopy measurements in THz spectral domain. The excitation source was a continuous semiconducting laser operating at $\lambda=645$ nm with output power of 40 mW. Laser radiation was chopped with frequency of 75 Hz. It was sent through a set of filters and diaphragms preventing penetration of the blackbody background radiation at THz frequencies from laser itself. THz emission from the sample was analyzed in collinear geometry with the excitation beam. Spectral measurements were performed using a step-scan Fourier spectrometer for the 5–350 cm^{-1} spectral range. The internal volume of the spectrometer was evacuated down to the residual pressure level of 6×10^{-2} Torr to minimize water vapor absorption. The THz radiation was detected by a liquid helium cooled Si bolometer whose output signal was measured by a lock-in amplifier.

Figure 2 shows the typical spectrum of THz emission emanating from undoped GaAs layer subject to interband photoexcitation. The PL peak is centered at ~7.2 meV that is slightly above the binding energy of shallow donor in GaAs of 5.9 ± 0.1 meV (see, e.g., Ref. 10). Taking into account the precise calibration of our detector with the emis-

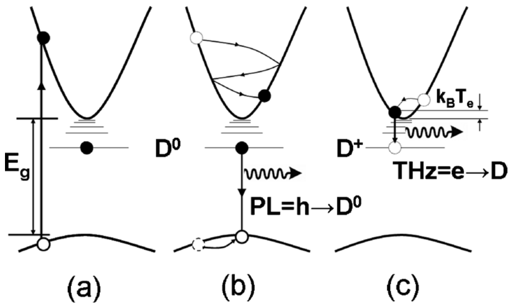

FIG. 1. Scheme of recombination processes in n-doped GaAs at band-to-band photoexcitation leading to THz PL: (a) optical excitation of (e)–(h) pairs, (b) relaxation of nonequilibrium carriers and recombination of free holes with electrons of neutral donors, and (c) THz radiative transitions of nonequilibrium electrons contributing to binding to ionized donors.

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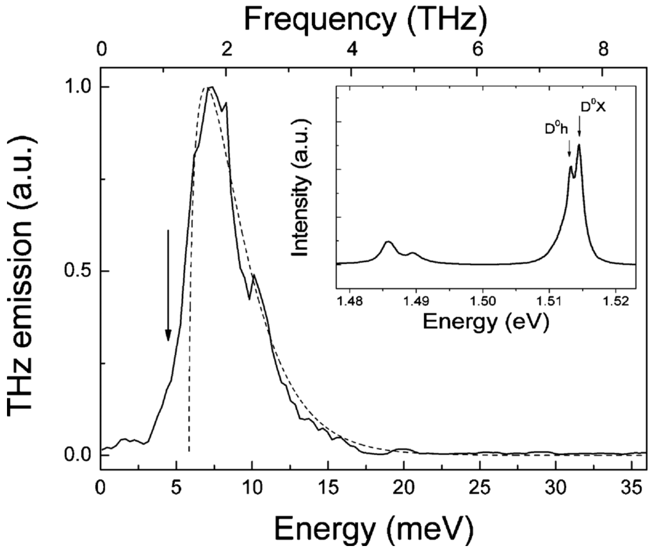

FIG. 2. Typical THz PL spectrum (solid curve) for undoped GaAs layer ($N_D-N_A\sim5\times10^{15}$ cm^{-3}). $T=5$ K. Excitation −645 nm line of semiconducting laser. $I_{exc}\sim16$ W/cm^2. The dashed line is calculated emission spectrum for optical transitions of electrons with $T_e\sim25$ K from the conduction band continuum to the donor ground state 1S. The arrow points to the energy of 4.4 meV corresponding to 2P→1S intracenter optical transitions for a shallow donor in GaAs (Ref. 2). The figure inset shows the spectra of near band edge PL of these samples excited by 645 nm line of semiconducting laser. $T=4.2$ K, $I_{exc}\sim2$ W/cm^2. Arrows point to the D^0X (recombination of exciton bound to neutral donor) and D^0h (free-to-bound recombination) emission lines, at 1.514 eV and 1.513 eV, respectively, identified in accordance with published data (see, for example, Ref. 8).

sion of a black body source and measured attenuation factor of the experimental instrumentation, we found the value of order of 10^{-6} for the spectrally integrated power efficiency of the THz source. Recalculation of the power efficiency to the external quantum yield η_{ext} of the luminescence with using the quantum energies of the excitation and THz photons gives the value $\eta_{ext}\approx2\times10^{-4}$ for the undoped GaAs layers. Results of a control experiment on detection of the THz PL in anticollinear geometry with the excitation beam show that the observed emission spectrum (Fig. 2) is undistorted by lattice absorption in GaAs.

We attribute the observed THz PL (Fig. 2) to optical transitions of nonequilibrium electrons from the conduction band to charged donor centers, created as a result of the D^0h recombination in accordance with the scheme outlined in Fig. 1. The D^0h emission line is clearly seen in the spectrum of near band edge PL (see Fig. 2 inset). The measurable high energy shift of the THz emission maximum from the donor binding energy can be explained by electron heating effects. Our estimations show that the effective temperature T_e of electrons in the conduction band can reach up to 20–30 K within conditions of the experiment. One can see from Fig. 2 that the THz emission spectrum is well consisted with the calculated emission spectrum for optical transitions of electrons with $T_e\sim25$ K from the conduction band to the donor ground state 1S. This calculated spectrum was obtained using the model of radiative transitions of electrons from continuum to discrete states of a hydrogen atom,[11] modified taking into account the donor binding energy. In accordance with the model, the optical transitions to excited impurity states are less significant because of the transition probability is proportional to ε^4, where ε is the energy of the transition. The intracenter optical transitions of the 2P→1S type presumably contribute to the THz emission also but due to considerable inhomogeneous broadening they are not clearly resolved and can be seen only as a relatively weak low-energy shoulder of the main emission band (see Fig. 2). The long arrow on Fig. 2 corresponds to the energy of 4.4 meV known for the transitions between the 2P and 1S levels of a shallow donor in GaAs.[2]

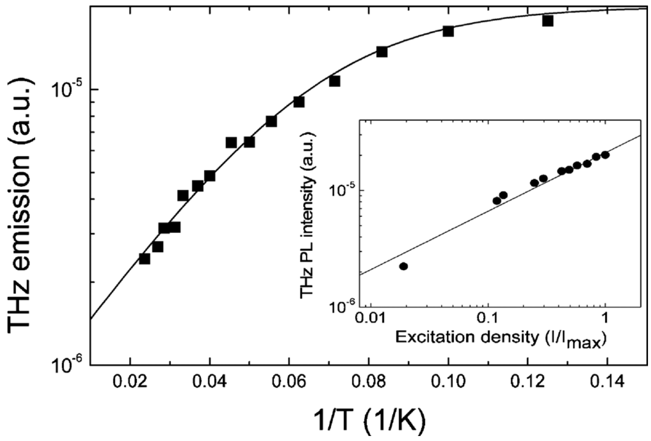

FIG. 3. Temperature dependence of the spectrally integrated intensity of the THz PL from undoped GaAs layer. $I_{exc}\sim16$ W/cm^2. Points are experimental data. The solid curve shows the least square fit of the experimental data by equation $I_{THz}(T)=(I(0)/1+D\exp(-E_q/k_BT))$ with $D=20\pm0.1$ and $E_q=4.0\pm0.1$ meV. The figure inset shows the dependence of the spectrally integrated THz PL intensity on the excitation density for undoped GaAs layer at 5 K. Points are experimental data. Curve is the result of approximation of the data by $I_{THz}=\text{constant}\times\sqrt{I_{exc}}$ law.

The intensity of THz emission strongly decreases with an increase in temperatures (Fig. 3). At temperatures above 50 K, THz PL becomes hardly detectable. This temperature quenching is presumably associated with several factors, among them thermal ionization of donor centers, as well as a decrease in probability of the capture of electrons by the donor centers with an increase in temperature. But we did not observe essential temperature-dependent modifications of the THz PL spectra. The Arrhenius plot delivers the quenching energy of 4.0 ± 0.1 meV (see Fig. 3), which by the order of magnitude agrees with the donor binding energy. The inset of Fig. 3 shows the plot of THz intensity dependence, I_{THz}, on excitation density, I_{exc}, that demonstrates a clear sublinear character. At the high excitation densities this dependence can be well approximated by $I_{THz}-I_{exc}^{\beta}$ law, where $\beta=0.5$. Within the framework of the model Fig. 1, we have calculated the intensity of THz generation using the set of rate equations,[12] describing the generation-recombination processes of nonequilibrium carriers created by interband photoexcitation in a weakly compensated n-GaAs. Results of these calculations give the linear dependence $I_{THz}-I_{exc}$ or the square root dependence $I_{THz}-\sqrt{I_{exc}}$ in the cases of low and high excitation densities, respectively, that is in reasonable agreement with the experimental data.

Figure 4 shows the spectrum of THz emission from Si doped GaAs layer with $N_D-N_A\sim8\times10^{16}$ cm^{-3} (curve 1). The emission intensity is increased by more than one order of magnitude comparing with the undoped GaAs layer (curve 2). The external quantum yield of THz PL from Si doped GaAs reaches the value of $\sim3\times10^{-3}$. The primary maximum of the PL is ~6.5 meV, but the overall character of the emission spectrum remains the same. In this case, the

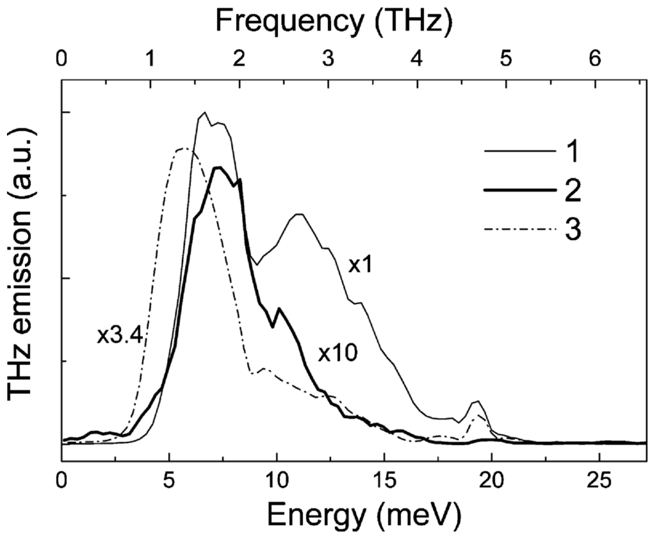

FIG. 4. Spectra of THz PL from Si-doped n-GaAs layer with $N_D-N_A \sim 8 \times 10^{16}$ cm^{-3} (curve 1), undoped GaAs layer with $N_D-N_A \sim 5 \times 10^{15}$ cm^{-3} (curve 2) and Si-GaAs substrate sample (curve 3). $T=5$ K, $I_{exc} \sim 16$ W/cm^2.

main THz emission can be attributed to optical transitions of the nonequilibrium electrons from the conduction band to empty states in the impurity subband, since these samples have delocalized donor states. Such additional empty states in the impurity subband appear as a result of recombination of nonequilibrium holes with the electrons of the impurity subband. One can see also THz PL peaks at ~11 and 19.3 meV. The weak resonance at 19.3 meV can be tentatively attributed to the radiative transitions of the holes to charged minor acceptors, which are present in the crystal. It can be either optical transitions from the valence band continuum to ground impurity state or intracenter optical transitions. Although the radiative mechanism responsible for the feature at 11 meV cannot be firmly established at this stage, it is present in all studied samples and is likely due to residual extrinsic defect in GaAs and requires more detailed specific research of defect formation. The THz PL was observed also from the test sample of semi-isolating GaAs (the substrate sample), whose typical spectrum is shown in Fig. 4 (curve 3) for comparison. We attribute the main THz emission from semi-isolating GaAs also to optical transitions of nonequilibrium electrons from the conduction band to the ground state of ionized donors. The ionized donors exist originally in such a heavily compensated material as semi-isolating GaAs is. The emission maximum is at ~5.7 meV (see Fig. 4), that is somewhat smaller comparing with the THz emission peak of n-GaAs samples. Such a decrease in the emission energy can be caused by decrease in binding energy of the donor centers due to screening of donor Coulomb potential by charged impurities existing in high quantity in the compensated material. Weak THz emission is seen also at the energies of ~10–11 meV and 19.3 meV already mentioned above.

To summarize, the efficient THz emission from n-GaAs under continuous-wave band-to-band photoexcitation at helium temperatures has been found out. We attribute the main PL to the radiative trapping of nonequilibrium electrons either to ionized donor centers (in the case of localized impurity states), or to empty states in the donor impurity subband (in the case of high impurity concentration, when the impurity subband is formed). The ionized donors or empty states in the impurity subband can be created in crystals as a result of impurity assisted electron-hole recombination. In compensated material the THz PL can appear due to the direct trapping of nonequilibrium carriers to charged impurity centers, which originally exist in the crystal. The external quantum yield of the THz emission has approached ~0.3% that makes the discovered phenomenon promising for practical applications.

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[1] S. H. Koenig and R. D. Brown, Phys. Rev. Lett. **4**, 170 (1960).
[2] I. Melngailis, G. E. Stillman, J. O. Dimmock, and C. M. Wolf, Phys. Rev. Lett. **23**, 1111 (1969).
[3] T. N. Adam, R. T. Troeger, S. K. Ray, P.-C. Lv, and J. Kolodzey, Appl. Phys. Lett. **83**, 1713 (2003).
[4] A. V. Andrianov, A. O. Zakhar'in, I. N. Yassievuch, and N. N. Zinov'ev, JETP Lett. **79**, 365 (2004).
[5] M. S. Kagan, I. V. Altukhov, E. G. Chirkova, V. P. Sinis, R. T. Troeger, S. K. Ray, and J. Kolodzey, Phys. Status Solidi B **235**, 293 (2003).
[6] V. A. Shalygin, L. E. Vorobjev, D. A. Firsov, V. Yu. Panevin, A. N. Sofronov, A. V. Andrianov, A. O. Zakhar'in, A. Yu. Egorov, A. G. Gladyshev, O. V. Bondarenko, V. M. Ustinov, D. V. Kozlov, and N. N. Zinov'ev, Appl. Phys. Lett. **90**, 161128 (2007).
[7] S. G. Pavlov, H.-W. Hübers, E. E. Orlova, R. Kh. Zhukavin, H. Riemann, H. Nakata, and V. N. Shastin, Phys. Status Solidi B **235**, 126 (2003).
[8] A. V. Akimov, A. A. Kaplyanski, V. V. Krivolapchuk, and E. S. Moskalenko, JETP Lett. **46**, 42 (1987).
[9] M. Benzaquen, D. Walsh, and K. Mazuruk, Phys. Rev. B **36**, 4748 (1987).
[10] R. J. Almassy, D. C. Reynolds, C. W. Litton, K. K. Bajaj, and G. L. McCoy, Solid State Commun. **38**, 1053 (1981).
[11] V. A. Fock, *The Principles of Quantum Mechanics* (Nauka, Moscow, 1976); *The Principles of Quantum Mechanics* (Mir, Moscow, 1978).
[12] M. Balkanski and R. F. Wallis, *Semiconductor Physics and Applications* (Oxford University Press, New York, 2000).