

Nitride-based photodetectors containing quantum wells in tunable electric fields*

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In this study, we present the optical properties (electroreflectance, luminescence and photosensitivity) of nitride nanostructures containing quantum wells (QW) inside GaN/AlGaN cavities designed so that the electric field inside them could be changed. The cavities contained one InGaN QW or two GaN QWs. We have confirmed experimentally that the electric field, controlled by external bias or by optical pumping, could change the properties of the structure. For example, (i) due to the Stark effect, a photoluminescence peak shifted from 2.97 eV to 3.06 eV, when the bias changed from +0.8 V to -2 V, (ii) due to various directions of the electric field inside the structure, the photocurrent changed not only in its value but also in its direction. The structures had non-linear photosensitivity. A double photoexcitation experiment showed that light from a second source could cause amplification or attenuation of the PC signal. It is proposed that the structures can be used to build active photodetectors which change their photoresponse in reaction to an external voltage or to illumination from another source.

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1. Introduction

Gallium nitride and related semiconductor compounds are wide band-gap materials that are used for the production of blue and UV light emitting diodes, lasers and photodetectors [1,2]. Moreover, nitrides are chemically robust and non-toxic against living cells, so they are used also as chemical and biological sensors [3]. For example, potassium selective chemical sensors [4] and prostate specific antigen detectors [5] using AlGaIn/GaN high electron mobility transistors have been proposed.

Since nitrides in a wurtzite structure exhibit a large spontaneous and piezoelectric polarization [6, 7], it is relatively easy to obtain a high electric field in the nitride structures. The spontaneous polarization can create high concentrations of carriers without doping [8], which is important for HEMTs [9] and other devices [10]. Moreover, the strong electric field can change the electron energies, tunnelling probabilities through barriers and carrier recombination rates. The field influence is important during photoexcitation of a semiconductor, when electrons and holes are generated simultaneously and then separated by the field. Photodetectors that benefit from this effect have been reported, including GaN/AlGaIn detectors with a spectral response tunable by external voltage [10,11].

In this paper, we describe a GaN/AlGaIn structure with a cavity designed so that the electric field inside could be changed by an external voltage or by illumination. The cavities contained quantum wells, so it was possible to investigate the properties of the quantum well (QW) in a significant electric field. Numerical modelling [10] made for these structures showed that illumination could change the carrier distributions and consequently change the field and potential. When under illumination, photo-injected electrons and holes formed a dipole that changed the field. Moreover, due to the different dynamics of electrons and holes, some electric charge could be accumulated, which changed the potential. Photo-excitation of barriers led to accumulation of electrons (holes drifted to the surface), which decreased the potential in the cavity. In this way, illumination and an electric field caused significant changes in the electrical and optical properties of the photodetectors.

2. Preparation and characterisation of the structures

The samples were grown by metal-organic chemical vapour deposition (MOCVD) on 2-inch-diameter sapphire substrates. On a thick (2-3 μm), Si-doped buffer layer,

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there were grown several thin (tens of nm), intentionally undoped layers. The layers formed three most important parts of the nanostructure: a deep barrier, a cavity with QWs and a top barrier. The whole structure was covered with a very thin GaN cap (about 2 nm).

Two types of nanostructure were investigated. The first type had only one $\text{Ga}_{0.9}\text{In}_{0.1}\text{N}$ quantum well (the energy band gap of this alloy being $E_g = 3.15$ eV [12,13]). The second type had two GaN QWs ($E_g = 3.42$ eV) and a higher aluminium content in the buffer and the barriers. The cavity layers were made of 24 nm $\text{Al}_{0.05}\text{Ga}_{0.95}\text{N}$ and 40-nm $\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}$ in types 1 and 2, respectively (see Fig. 1). Without any strain, these alloys would have band-gaps of 3.52 eV and 3.63 eV [12,13].

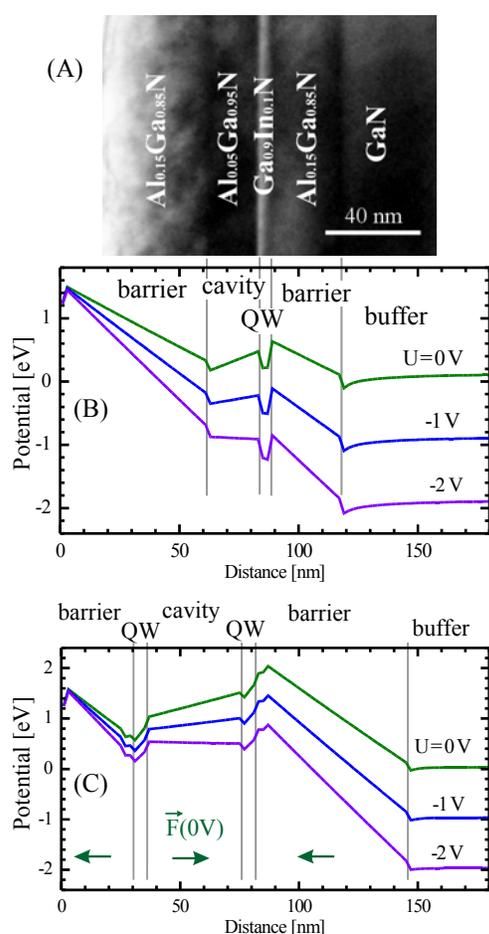


Fig. 1. A) TEM picture of the type 1 sample, B) the potential distribution obtained by modelling. C) The potential distributions for the type 2 structure (the arrows show the directions of the electric field).

The barriers had compositions $\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$ and $\text{Al}_{0.2}\text{Ga}_{0.8}\text{N}$ in types 1 and 2 respectively, so in the case of the full relaxation their energy gaps would be 0.2 eV wider than the cavity energy gaps. The barriers had the following widths: sample type 1 - deep barrier 30 nm, top barrier 60

nm, type 2 - deep barrier 60 nm, top barrier 30 nm (see Fig. 1).

In fact, the potential in a semiconductor nanostructure depends not only on the composition but also on the electric field. The field is created by external bias, electric charge inside the structure and by piezoelectric and spontaneous polarisation in the layers. An electric field F acting along the z axis adds a potential $V_q = e \int F dz$ which can be calculated by a simple procedure [10]. The resulting potential at the bottom of the conduction band is plotted in Figs. 1B and 1C. At the base of Fig. 1C, the directions of the electric field were plotted. It was found that (at zero bias and without illumination) the electric field in the cavity had an opposite direction than the field in the barriers. The field generated by the spontaneous polarisation has a direction to the surface (negative). Its value increases with increased aluminium content. The charge transferred by the polarisation-related field generated an electric field of reversed (positive) direction. Since, in the low-aluminium cavity, the polarisation-related field was weak, the charge-related field dominated in the cavity.

The widths of the QWs varied with the position on the wafer, which gave us a possibility to choose samples with different QW widths. Transmission electron microscopy (see Fig. 1A) and capacitance-voltage measurements helped us to determine the widths of the QWs and the whole structure. In the type 1 wafer, the QW's width was between 2 and 3 nm. In the type 2 wafers, the QW widths were between 4 and 6 nm.

Semitransparent Schottky contacts were made by the evaporation of gold. Melting indium into a side of the sample made ohmic contacts to the conductive buffer (resistance below 1 k Ω). The current-voltage characteristics showed good electrical properties of the investigated devices.

Electroreflectance (ER) spectroscopy was used to determine the electric fields in the detector structures (see Fig. 2). The shape of the ER signal from a quantum well hardly depended on the electric field, so we could determine only the energies of the features at about 3.1 eV and 3.5 eV for the GaInN QW and the GaN QW, respectively. The lines from the cavity changed significantly with bias (see Fig. 2). However, their behaviour was difficult to interpret. In contrast to this, the signal from the AlGaIn barriers showed clear

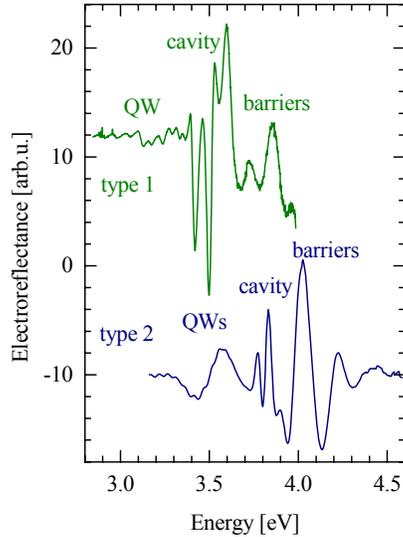


Fig. 2. Electroreflectance spectra of the two types of sample. The Franz-Keldysh oscillations are most visible for the barriers of the type 2 sample.

Franz-Keldysh oscillations that were used for the calculation of the electric field [14].

It was found that the field in the barriers was high, even without bias: 0.25 MV/cm, 0.4 MV/cm, for samples of types 1 and 2, respectively. The field was much higher at a high negative voltage, of about 1 MV/cm. It increased linearly with the bias voltage of 0.09 MV/cm and 0.17 MV/cm per 1 V, for the types 1 and 2 samples, respectively. The experimental values of the field in the barriers were similar to the field obtained by numerical modelling. This suggested that we could rely on the calculation results also in the case of the cavities and the quantum wells.

3. The QWs in an electric field - results and discussion

The carriers excited by illumination can either recombine or drift in the electric field creating a photocurrent (PC). In our samples, at low temperatures, the photoexcited carriers had a high probability of radiative recombination. Thus, it was possible to get important information by photoluminescence (PL) measurements. At high temperatures, carriers had a high probability of jumping over the barriers and it was possible to measure the photocurrent. The results of the PL and PC measurements of the structure are presented below.

3.1. Photoluminescence measurements

The photoluminescence was measured at room temperature and at the liquid helium temperature. It was excited by a He-Cd laser $\lambda = 325$ nm ($h\nu = 3.81$ eV) and measured with a 0.5-m spectrometer. The excitation power density was in the range $10^{-3} - 10^{-4}$ W/cm². The excitation energy was too low to reveal luminescence from the

barriers, but it was sufficient for measurements of the QWs.

In samples with a GaN buffer (type 1), the GaN line was observed at 3.418 eV at room temperature and 3.481 eV at 4 K. These are standard values for GaN grown on sapphire [15].

At room temperature, the QW luminescence was broad (about 0.1 eV). Its energy was between 3.0 and 3.2 eV. The energy changed from sample to sample, depending on the position on the wafer. The PL was stronger in the type 1 samples than in the type 2 samples. Its amplitude was proportional to the power squared, which was due to fast non-radiative recombination or to escape of the carriers from the QWs.

At low temperature (4 K), the PL from the QWs was easily visible (see Fig. 3). The QW PL peaks had widths of about 40 meV.

Phonon replicas of the QW luminescence were observed at energies shifted by 84 - 88 meV (they are most visible in Fig. 4A). This value is well between the energies of the LO phonon in GaN (92 meV) and InN (73 meV) [16], so we concluded that these were LO phonon replicas (QW,LO).

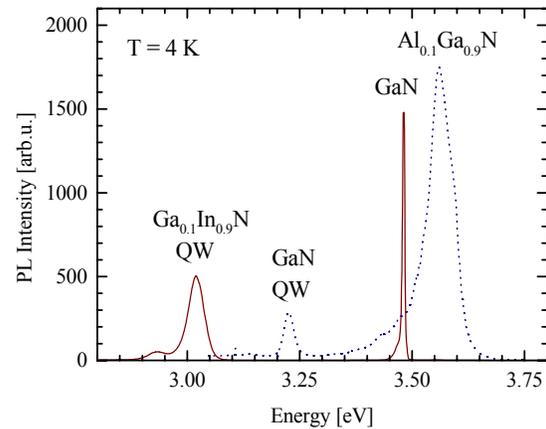


Fig. 3. PL spectra of two samples: type 1 with a 3-nm GaInN QW (solid line) and type 2 with 4-nm GaN QWs (dashed line).

As mentioned in Section 2, the width of a QW varied depending on the position in the wafer. In this Section, we focused on two samples: type 1 with a 3-nm GaInN QW and type 2 with 4-nm GaN QWs. The PL energies measured outside a Schottky contact were about 3.03 eV and 3.23 eV for the 3-nm Ga_{0.1}In_{0.9}N QW and for the 4-nm GaN QWs, respectively. Both values were smaller than expected for a simple, unperturbed, QW.

Taking into account a rectangular QW of width L with barriers of infinite height, one can expect that the electron energy would be increased by the quantisation energy:

$$W_{\text{QW}} = \frac{h^2}{8L^2 m^*}, \quad (1)$$

where h is the Planck constant and m^* is the effective

mass of an electron. Based on Eq. (1), the expected energy W_{QW} should be 0.24 and 0.13 eV for the two described types of QW.

More exact calculations, taking into account the finite barrier heights, gave smaller quantisation energies, W_{QW} . So, for the nominal composition of the QWs we expected the PL energies: 3.25 and 3.48 eV - still higher than the experimental values. The observed differences could be due to a lower than nominal In content (in the case of the GaInN QW) or due to the quantum confined Stark effect. When an electron is confined in a QW, it cannot drift in an electric field F . Instead; its wave function $\psi(z)$ is moved by the field to the wall of the QW and squeezed. The Schrödinger equation should be completed with a potential $V = -eFz$. In the case of a rectangular QW of width L , with infinite barriers, the solution is a sum of the Airy functions [17]. The resulting energy eigenvalue is lower by an energy of the order of eFL (about 0.1 eV).

The calculated dependence was nonlinear. In the range $eFL < 10 \cdot W_{QW}$, it can be approximated as:

$$E = E_g + W_{QW} \left(1 - \alpha \frac{eFL}{W_{QW}} - \beta \frac{(eFL)^2}{W_{QW}^2} - \dots \right), \quad (2)$$

where E_g is the alloy energy gap, $\alpha = 0.0090$, $\beta = 0.0086$.

In order to investigate the effect of the electric field, we applied a voltage to one of the diodes and measured the photoluminescence (see Fig 4.).

The PL spectra showed that the electric field changed the energy of the QW peak. In one of the type 1 samples, the QW peak shifted from 2.97 eV ($U = +0.8$ V) to 3.06 eV ($U = -2$ V) (see Fig 4).

As described in Section 2, due to the spontaneous polarisation and the field generated by the electric charge, the field in the cavity had a positive direction, so that positive bias made it stronger. On the other hand, a negative voltage introduced a field opposite to the build-in field, so the field decreased. At about -1 V, on a high energy wing of the PL peak, a second peak appeared. This peak was related to a higher electron state in the QW. Its appearance suggested that the occupation of the QW increased significantly. This could be explained by the illumination-related decrease of the potential around the QW. Unfortunately, the higher concentration of electrons strongly screened the external (bias-related) electric field, and so further changes of the field were not possible.

Since the negative voltage introduced a field opposite to the build-in one, the field in the cavity decreased. In a low field, the electron wave function occupies the whole QW - the parts with low and with high potentials, so the electron energy was only slightly sensitive to the potential generated by the electric field (see Fig. 4).

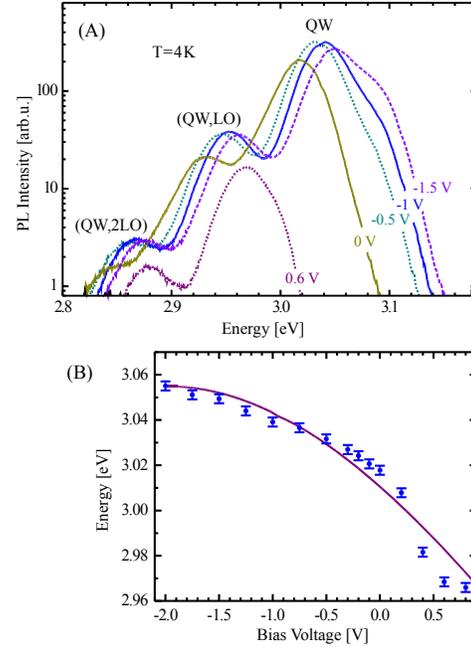


Fig. 4. A) Photoluminescence spectra at different bias voltages. B) Energy of the QW peak versus voltage (points) and the calculated energy change in the electric field (solid lines).

The positive voltage increased the electric field in the cavity. The electron wave function was moved by the field to the wall of the QW and squeezed. The whole function was in a low potential generated by the electric field, so the electron energy decreased quickly with the field.

The curve given by Eq. (2) was fitted to the experimental data (see Fig. 4B). The fitting parameters $E_g = 3.96 \pm 0.05$ eV and $W_{QW} = 0.1 \pm 0.05$ eV were strongly correlated, so they could not give precise information. However, it was obvious that the QW behaved in agreement with the Stark effect predictions. The lower than expected value of E_g was probably due a higher than nominal indium content in the QW. The quality of the fit could have been improved, by taking into account changes of energy of the holes, but it would add too many free parameters to the fit.

3.2. Photocurrent measurements

Photoelectric properties of the samples were measured with the use of a logarithmic picoammeter (sensitivity 0.1 pA). The photocurrent was excited by light from a halogen incandescent lamp, filtered by a 320-cm monochromator. The samples were measured in the energy range 0.6 to 4.2 eV. Generally, they were practically insensitive to radiation of energies below 3 eV. Therefore, most of the measurements were taken in the range 3.0 – 4.2 eV.

Current-voltage (I-V) characteristics showed good electrical properties of the investigated devices. The shape of the I-V characteristic measured in the darkn was similar to a diode I-V characteristic. At $U = -1$ V, the dark current density was of the order of 1 nA/cm², and the photocurrent efficiency was of the order of 0.1 A/W. Both parameters

were higher in the type 1 samples and lower in the type 2 samples.

The current-voltage characteristics were measured under different illuminations (see Fig. 5). Under reverse bias, the light caused a significant increase in the current and changed the shape of the characteristics to transistor-like ones.

It could be observed that the I-V characteristics had different shapes, depending on the layer that absorbed the light. In the case of light absorbed by barriers, a significant current was generated. An increase in the current was easily observed at reverse bias and at low positive bias ($U < 1$ V). At higher bias, the dark current (that increased exponentially with voltage) was too strong for the PC measurements. On the other hand, in the case of light absorbed by the QWs, the current was much weaker. In the range above some threshold voltage $U_T = -0.8$ V (type 1 sample) or $U_T = -1.8$ V (type 2), the I-V characteristics were very similar to the dark I-V characteristics. Only below the U_T did the device open and a significant PC was generated (see Fig. 5).

A similar effect was already reported for AlGaIn/GaN structures [10,11] and explained by the influence of barriers formed by the valence band offset and the spontaneous polarization of the $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_y\text{Ga}_{1-y}\text{N}$ interface. The photoexcited carriers are stopped and accumulated at the interface. A 2D electron gas is formed on the interfaces (electrons are attracted by the electric field caused by spontaneous polarization). The photogenerated holes recombine with the electrons, and the photocurrent is stopped.

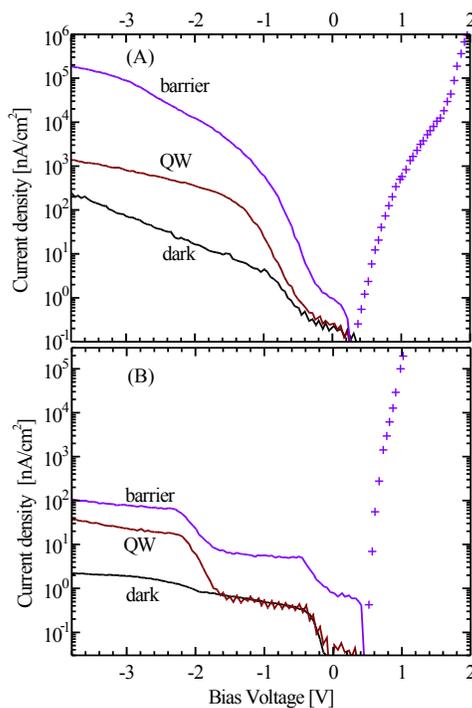


Fig. 5. Current - voltage (I-V) characteristics measured under different illuminations. A) The type 1 structure. B) The type 2 structure.

Numerical calculations of the electric field and the potential profiles (see Fig. 1) were made for the investigated structures. The obtained distributions of the electrons and the electric field showed that at the bias voltage U_T the electric field in the cavity was reduced to zero, and then changed its sign. So, below the threshold U_T , the 2D electron gas at the cavity-barrier interface could not exist and the device was opened for the PC generated in the QW. The effect was much more visible in the type 2 samples, where the cavities were broader.

During the photocurrent measurements the light was modulated, so the PC was measured as a difference of the current measured during illumination and the dark current. The modulation frequency was about 0.1 Hz.

The photocurrent spectra showed the presence of bands related to all AlGaIn layers, and to the QWs. The barrier-related band weakly depended on the bias voltage. On the other hand, the low energy PC-band (related to the cavity and the QW) changed strongly with bias voltage. It changed not only its magnitude but also its sign. The ratio between the highest and lowest significant PC signal was about 10^5 . Since the current value was in some spectral range negative, but in another positive, it was impossible to use a logarithmic scale, so the PC spectra were plotted in a cubic root ($I^{1/3}$) scale (see Fig. 6). Here, the positive photocurrent means a current going from the Schottky contact to the ohmic contact.

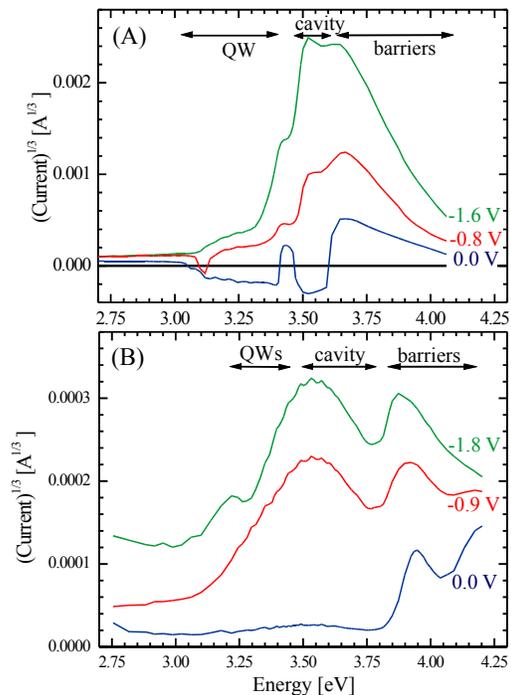


Fig. 6. The photocurrent spectra measured under different bias voltages for A) the type 1 and B) the type 2 sample.

The PC had different signs, depending on the photon energy. The effect was easily visible on the PC spectra of

the type 1 samples plotted in Fig. 6A. The PC was negative at $h\nu = 3.2$ eV (QW excitation), positive at 3.42 eV (the GaN buffer), negative at 3.5 eV (the cavity) and back - positive at 3.7 eV (the barrier). Since the sign of the PC depends on the direction of the electric field in the place where electron-hole pair is excited, we can conclude that the direction of electric field in the cavity was opposite to the field in the barriers, in agreement with the model presented in Fig. 1.

When the cavity was excited for too long (10 s or more at a power of 0.1 μ W), the photogenerated charge carriers accumulated at the opposite walls of the cavity, and screened the electric field. When the field was reduced, the negative photocurrent was switched off.

For type 2 samples, the negative photocurrent was practically not observed. At zero or low reverse bias ($U > -0.5$ V), the PC signal from the QWs was practically absent. The current blockade above U_T was much more efficient in type 2 samples, because of the broader cavity layer. Under a strong ($U < U_T$) reverse bias, it increased by about 3 orders of magnitude (see Fig. 6B). As mentioned above, the effect was related to the reduction of the electric field and the depletion of the electron concentration at the cavity-barrier interface.

4. The AlGaIn structure as an active photodetector

As was shown in Section 3, the optical properties of the structure could be changed by illumination. For example, an increase of the excitation power caused a change of shape and an increase of the energy of the PL peak.

Also, in the case of the photoelectric measurements, significant changes in the PC spectra, resulting from the changes of the excitation power, were observed (for both types of sample).

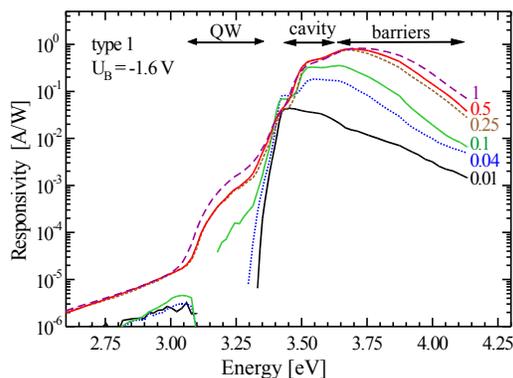


Fig. 7. Power dependence of the photocurrent. The spectra were divided by excitation powers, to show the relative change of the photosensitivity.

The PC spectra of a type 1 sample were divided by excitation power, to show relative change of the photosensitivity, and are plotted in Fig. 7. Units of the

photon flux, as used on this figure, were of the order of $3 \cdot 10^{10}$ photons/s (20 nW).

Assuming that one photon excites one electron, the photocurrent should be proportional to the flux of photons or to the excitation power, which is in fact observed for standard photodiodes. Since the photocurrent spectra presented on Fig. 7 are divided by the excitation power they should have the identical shape - independent of the illumination. However, it was observed that the relative signal increased with the power. This means that absolute PC signal increased faster than linearly. In fact, for low excitation, the PC had a squared dependence for the spectral range above 3.5 eV (excitation of the cavity and the barriers). The excitation of the QW (3.0–3.3 eV) had a more complicated dependence. For low excitation, the relative PC signal was negative, and for stronger excitation it became positive. For excitation powers higher than 0.25 (Fig. 7) the relative signal saturated, which means that the absolute PC signal increased linearly.

The structures presented here changed their properties after illumination, so they could be used to build detectors with spectral responses tuned by the illumination. When such a device is exposed to two sources of illumination, its response depends on both input signals. The input signals can be of different wavelengths.

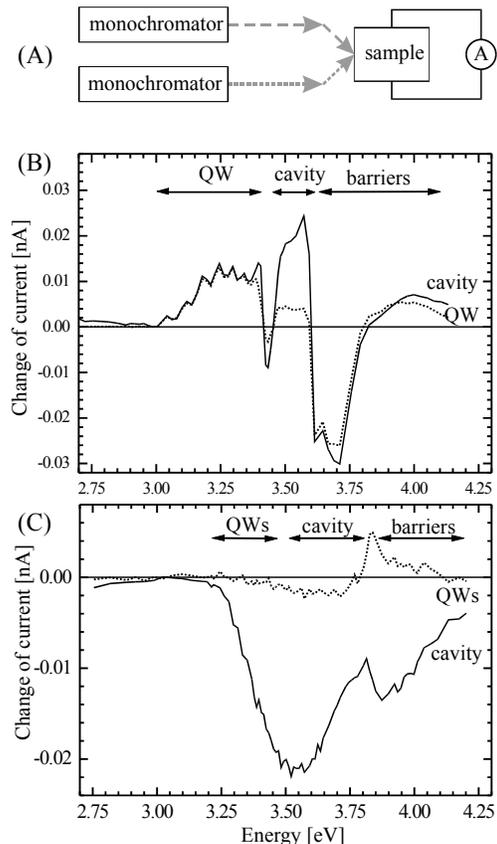


Fig. 8. A) Idea of the double photoexcitation experiment. B) and C) - spectra of type 1 and 2 samples. Solid line - change of the photocurrent spectrum caused by the excitation of the cavity, dotted line - the excitation of the QW.

In order to check this possibility, we conducted photocurrent measurements with two excitation sources. The scheme of the double photoexcitation experiment (DPE) is plotted in Fig. 8A. The samples were biased by a voltage equal to the threshold bias, U_T , observed in the I-V characteristics (see Section 3.2).

The spectra in Fig. 8 show changes of the photocurrent spectrum caused by the second excitation. If the photoelectric response was insensitive to the second excitation, the DPE signal should be zero. A positive signal means amplification of the sensitivity, and a negative one, attenuation.

In the type 1 samples, attenuation was observed only for narrow bands at 3.5 eV (the GaN buffer excitation) and 3.7 eV (the 2D electron gas). Amplification was obtained for the rest of the spectrum (including the QW, the cavity and the barriers). This was because the additional photoexcited carriers screened the electric field in the cavity and opened the device. In this case, when the two excitations came together, the output signal was much stronger. This could be interpreted as a conjunction (AND) of the two input signals. It would be similar to an electronic logical gate, but with optical inputs and an electric output.

On the other hand, the excitation of type 2 sample caused attenuation. In this situation, the device responded to the first input signal or to the second, but not to two simultaneous signals. This could be interpreted as exclusive disjunction (XOR) of the two signals. The attenuation was also caused by additional photoexcited carriers that screened the electric field in the cavity. However in this case, the electric field in the cavity had already a forward direction (the sample was biased), so the screening of the field caused a decrease in the photocurrent.

5. Conclusions

We have presented research on photodetector nanostructures built from nitride semiconductor alloys (GaIn)N and (AlGa)N. The structures contained cavities and quantum wells. Two types of sample were presented: an $\text{Al}_{0.05}\text{Ga}_{0.95}\text{N}/\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$ structure with a $\text{Ga}_{0.9}\text{In}_{0.1}\text{N}$ QW and an $\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}/\text{Al}_{0.2}\text{Ga}_{0.8}\text{N}$ structure with two GaN QWs. General comparison shows that the type 1 samples were more sensitive to light, and the type 2 samples had a lower leakage current. The most important differences were caused by the electric field inside them that changed their optical properties.

The presented devices profited from the piezoelectric and spontaneous polarizations generated on the GaN/AlGaN interface that created a strong electric field inside the semiconductor nanostructure. The field attracted electrons, forming a 2D electron gas at the interfaces, and changed the energy of the electronic states in the quantum well. This was confirmed by photoluminescence measurements. that showed the change of the QW energy.

Photocurrent measurements showed that the PC spectra were changed strongly by bias - not only their magnitude but also their sign. This effect was obviously related to the change in the electric field direction.

The designed barriers enabled accumulation of the charge carriers, and caused fast recombination of the holes and the electrons. The fast recombination switched off the photocurrent. Since accumulation of photoexcited charge carriers at the barriers changes the electric field, it is possible to change the electric field inside the device by optical excitation. This was confirmed experimentally, by observation of changes of the photoluminescence and the photocurrent spectra. Moreover, in the double photoexcitation experiment, we observed that light from the second source could cause amplification or attenuation of the PC signal.

Finally, it is pointed that the structure can be used to build active photodetectors, which change their response for one light source when they are switched by illumination from another source. When such a device is exposed to two sources of illumination, its response would depend on both input signals. The double photoexcitation experiment showed that the structures could perform AND and XOR functions.

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