Investigation of photoconductivity of individual InAs/GaAs(001) quantum dots by Scanning Near-field Optical Microscopy

D O Filatov¹, I A Kazantseva², N V Baidus³, A P Gorshkov¹,³ and V P Mishkin⁴

¹ Research and Education Center for Physics of Solid State Nanostructures, Lobachevskii State University of Nizhnii Novgorod, Nizhnii Novgorod, 603950, Russia
² Department of Physics, Lobachevskii State University of Nizhnii Novgorod, Nizhnii Novgorod, 603950, Russia
³ Research Institute for Physics and Technology, Lobachevskii State University of Nizhnii Novgorod, Nizhnii Novgorod, 603950, Russia
⁴ Institute of Physics and Chemistry, Ogaryov Mordovia State University, Saransk, Mordovia Republic 430005, Russia

* dmitry_filatov@inbox.ru

Abstract. The spatial distribution of the photocurrent in the input window plane of a GaAs-based p–i–n photodiode with embedded self-assembled InAs quantum dots (QDs) has been studied with the photoexcitation through a Scanning Near-field Optical Microscope (SNOM) probe at the emission wavelength greater than the intrinsic absorption edge of the host material (GaAs). The inhomogeneities related to the interband absorption in the individual InAs/GaAs(001) QDs have been observed in the photocurrent SNOM images. Thus, the possibility of imaging the individual InAs/GaAs(001) QDs in the photocurrent SNOM images with the lateral spatial resolution ~ 100 nm (of the same order of magnitude as the SNOM probe aperture size) has been demonstrated.

1. Introduction

Scanning Near-field Optical Microscopy (SNOM) is a method of Scanning Probe Microscopy destined for measuring the spatial distribution of various optical parameters of the solid surfaces with the nanometer-scale spatial resolution (much less than the wavelength of the probing light, thus overcoming the diffraction limit of the spatial resolution of the ordinary optical microscopy) [1]. Among the optical parameters mentioned above, there are optical transmittance, reflectance, luminescence, Raman scattering, etc. However, the photoconductivity (PC) of the semiconductor (and other) nanostructures under the local photoexcitation via a SNOM probe [2] is studied rarely as compared to other SNOM techniques. At the same time, the PC SNOM studies of the self-assembled semiconductor nanoislands and quantum dots (QDs) have not been reported until recently.

In Ref. [3], we have applied PC SNOM to study the photocurrent of a Si-based p^+–n photodiode (PD) with embedded self-assembled GeSi/Si(001) nanoislands as a function of the position of the SNOM probe in the plane of the input window of the PD. The features observed in the PC SNOM images have been attributed to the interband optical absorption in the GeSi nanoislands followed by the emission of the photoexcited holes from the nanoislands and the separation of the electron-hole
pairs in the electric field of the \( p^+-n \) junction. Thus, the possibility of imaging the individual GeSi nanoislands with the lateral spatial resolution \( \sim 100 \) nm (i.e. of the same order of magnitude as the aperture size of the SNOM probe used) has been demonstrated.

In the present paper, we report on the PC SNOM investigation of the GaAs-based \( p-i-n \) PD with embedded self-assembled InAs/GaAs(001) QDs. The goal of the present study was to explore the capabilities of PC SNOM in the imaging of individual InAs/GaAs(001) QDs, which are much smaller (about an order of magnitude) than the GeSi/Si(001) nanoislands studied in Ref. [3].

2. Experimental details

The \( p-i-n \) diode structure has been grown on an \( n^+-\)GaAs(001) substrate by Low Pressure Metal Organic Vapor Phase Epitaxy (LP MOVPE) using Aixtron® AIX 200RF growth setup. The gas pressure in the reactor was \( \sim 100 \) mbar. A schematic representation of the investigated diode is shown in Figure 1. The InAs QDs were grown on the surface of a \( \approx 350 \) nm thick intentionally undoped \( i^-\)GaAs layer (the background electron concentration was \( \sim 5\times10^{15} \) cm\(^{-3} \)) by self-assembling via Stranski-Krastanov mechanism at the substrate temperature \( T_g \approx 490 \) °C. The InAs QDs were capped by a \( \approx 10 \) nm thick \( i^-\)GaAs layer. Finally, a \( p^+-\)GaAs subcontact layer with the thickness \( d_c \approx 50 \) nm doped by C with the acceptor concentration \( \approx 3\times10^{18} \) cm\(^{-3} \) was deposited. The \( p^+-\)GaAs layer thickness \( d_c \) was selected so that the QDs were within the near-field region from the surface, i.e. \( d_c < \lambda_{ex}/n \), where \( \lambda_{ex} \approx 1 \) \( \mu \)m is the photoexcitation wavelength and \( n \approx 3.5 \) is the refractive index of GaAs. A calculated equilibrium band diagram of the \( p-i-n \) diode structure with InAs QDs at 300 K is presented in Figure 2a. The values of the band offsets at the InAs/GaAs(001) QD interface as well as the dependencies of the quantum confined electron and hole energies on the QD sizes were taken from Ref. [4]. The charging of the QDs by holes was neglected.

The morphology of the InAs QDs were examined by Atomic Force Microscopy (AFM) in ambient air using NT-MDT® Solver Pro™ instrument in the Semicontact™ mode on the satellite sample with the surface InAs QDs grown in the same conditions as the ones in the \( p-i-n \) diode structure. NT-MDT® NSG-11 probes with the tip curvature radius \( R_p < 10 \) nm were used. The AFM data were analyzed by NT-MDT® Nova™ Image Analysis software using Grain Analysis block.

The InAs QDs in the AFM images were identified by the threshold method, and the statistical analysis of the morphological parameters of the QDs has been performed. The morphology of the QDs was characterized by the following parameters: the height \( h \) (the origin being selected at the InAs wetting layer (WL) surface), the averaged base diameter \( D \) (at the level of 0.1\( h \)), and the surface density \( N_s \).

![Figure 1. Schematic representation of the experimental setup for the PC SNOM measurements.](image-url)
Figure 2. (a) Calculated equilibrium band diagram (300 K) of the GaAs-based $p-i-n$ structure with embedded InAs/GaAs(001) QDs for the PC SNOM investigations; (b) the mechanism of PC due to the interband optical absorption in the InAs QDs.

The optical properties of the $p-i-n$ diode structure were examined by the photoluminescence (PL) spectroscopy at 77 and 300 K using LOMO® MDR-23 grating monochromator. The PL was excited by a continuous wave (CW) He–Ne laser with $\lambda_{ex} \approx 632.8$ nm and the output power $P \approx 30$ mW and registered by the lock-on technique.

On the base of the $p-i-n$ structure, the cylindrical mesa PDs with the input windows in the upper Au Ohmic contacts were fabricated. The mesa diameter was $\approx 350 \mu$m, the window diameter was $\approx 200 \mu$m. The bottom Ohmic contact to the back side of the $n^+\text{-GaAs}$ substrate was alloyed from a Sn foil by a spark discharge. The photoelectric properties of the PDs were examined by the PC spectroscopy at 300 K using LOMO® MDR-2 grating monochromator with a 100-W halogen lamp as the photoexcitation source. The spectra of photosensitivity (PS) $S_{ph}$ were measured in the shortcut circuit (SC) photocurrent mode with the chopped photoexcitation (the chopper frequency was $\approx 180$ Hz) using Stanford Research® SR-530 lock-in detector and were normalized to the photoexcitation intensity spectrum.

The PC SNOM investigations were carried out in ambient air at 300 K using NT-MDT® Solver™ SNOM scanning head and controller with modulated photoexcitation. The schematic representation of the experimental setup is shown in Figure 1. Dilas® DMPO131-14 and DMPO155-14 single mode CW pigtail semiconductor laser diodes (LDs) with the emission wavelengths $\lambda_{ex} \approx 1310$ nm and $\approx 980$ nm, respectively were employed as the photoexcitation sources. Both values of $\lambda_{ex}$ were within the interband optical absorption band of the InAs QDs in the investigated PDs as follows from the PL and PC spectra (see Section 3 below). The nominal values of $P$ in the pigtailed were $\approx 1.5$ mW for both LDs. However, in the experiments $P$ was limited to 200 $\mu$W in order to avoid the damage of the SNOM probe aperture due to burning. The power supply for the LDs was provided by Dilas® PILOT™ 4-DC LD driver. The photoexcitation intensity was modulated by modulating the LD pumping current by an optron driven by a master oscillator with the frequency of $\approx 360$ Hz. The photoresponse of the PD was recorded in the SC photocurrent mode by the same SR-530 lock-in detector connected to the user input of Solver™ SNOM controller. The NT MDT® SNOM probes with the integrated tuning fork shear force sensors were used. The nominal aperture size was $\approx 100$ nm.

3. Results and discussion
An AFM image of the satellite sample with the surface InAs/GaAs(001) QDs is presented in Figure 3a. The histograms of distribution of the QDs in the AFM image by $D$ and $h$ are presented in Figures 3b and 3c, respectively. The surface QDs observed in the AFM image has a three-modal distribution both by $D$ and by $h$. Besides the coherent InAs QDs with $h = 4-9$ nm and $D = 10-20$ nm, the relaxed InGaAs clusters with $D = 60-120$ nm and $h = 20-40$ nm have been observed. The latter
arise as a result of coalescence of the coherent InAs QDs during growth [5]. In turn, the coherent InAs QDs have a bimodal distribution by $D$ and $h$ typical for the InAs/GaAs(001) QDs grown by MOVPE [6]. The smaller QDs have $D = 8-12$ nm and $h = 4-5$ nm, while the bigger ones have $D = 16-20$ nm and $h = 6-9$ nm. It is worth noting that the lateral sizes of the InAs QDs in Figure 3 were enlarged as compared to the actual ones up to 40-50 nm due to the convolution effect [7].

The PL spectra of the $p-i-n$ structure with embedded InAs QDs at 77 and 300 K are presented in Figure 4a. Besides the peaks of GaAs edge PL at the photon energies $h\nu \approx 1.51$ and 1.42 eV at 77 and 300 K, respectively, the peaks at $h\nu \approx 1.07$ eV and $\approx 1.25$ eV related to the interband radiative optical transitions between the ground quantum confined electron and hole states in InAs QDs of bigger and smaller sizes, respectively were observed in the PL spectrum at 77 K. In the PL spectrum measured at 300 K, the PL peak from the bigger QDs (at $h\nu \approx 1.01$ eV) was observed only because the energy barrier for the electrons occupying the ground state in the QDs appears to be too small ($\approx 130$ meV according to [4]). It is comparable to the thermal energy $kT \approx 26$ meV at 300 K, so that the thermal emission of the excess electrons from the QDs prevails the radiative recombination inside the QDs [8]. As a result, the PL peak from the smaller QDs was not observed at 300 K.

In the PS spectrum (300 K) shown in Figure 4b, besides the PS band with the edges at $h\nu \approx 1.42$ eV and $\approx 1.35$ eV related to the GaAs intrinsic PS edge and to the InAs WL, respectively, the PS bands with the edges at $h\nu \approx 1.2$ eV and $\approx 0.95$ eV attributed to the PS of the InAs QDs of the smaller and the bigger sizes, respectively were observed. The PS of the QDs was related to the interband optical transitions in the QDs followed by the emission of the photoexcited carriers from the QDs into the GaAs host matrix and the separation of the electron-hole pairs by the electric field of the $p-i-n$ junction, as shown in Figure 2b.

The PS edge at $h\nu \approx 0.95$ eV was attributed to the relaxed InGaAs clusters. One can estimate the In molar fraction $x$ in the cluster material ($\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy) from the spectral position of the respective PS band edge as $x \approx 0.35$ [6]. It is worth noting that the PS bands of the bigger QDs and the InGaAs clusters in Figure 4b are $\sim 100$ times weaker than the one of the smaller InAs QDs. One of the reasons for this is a lower surface density of the bigger QDs than of the smaller ones ($\approx 2.2 \times 10^8$ cm$^{-2}$ and $\approx 4.9 \times 10^8$ cm$^{-2}$, respectively). Another reason is the recombination of the photoexcited carriers inside the bigger QDs and InGaAs clusters prevailing over the emission from these ones. The observation of the PL from the bigger QDs at 300 K supports the above explanation.

Note that the PL lines from the InGaAs clusters are not manifested in the PL spectra, because the relaxed clusters contain the misfit dislocations, so that the non-radiative recombination prevails over the radiative one.

**Figure 3.** (a) AFM image and the histograms of distribution of the InAs QDs and relaxed InGaAs nanoclusters (b) by base size $D$ and (c) by height $h$. 


Figure 4. (a) PL spectra (77 and 300 K) of the $p$–$i$–$n$ structure with InAs QDs; (b) PS spectrum (300 K) of the PD based on the $p$–$i$–$n$ structure. The dashed lines indicate the photon energies of the emission of the LDs employed in the PC SNOM measurements.

The PC SNOM images recorded at $\lambda_{ex} \approx 1310$ nm and $\approx 980$ nm are presented in Figures 5a and 5b, respectively. The inhomogeneities observed in the PC SNOM images were related to the interband optical transitions between the quantum confined electron and hole states in the InAs QDs as well as between the valence band states and the conduction band ones in the relaxed InGaAs nanoclusters. As it follows from the PC spectrum of the diode shown in Figure 4b, the biggest InAs QDs only as well as the InGaAs clusters can be manifested in the PC SNOM image in Figure 5a recorded at $\lambda_{ex} \approx 1310$ nm. In contrary, the smaller QDs can be manifested in the PC SNOM image recorded at $\lambda_{ex} \approx 980$ nm only.

The minimum size of the features observed in the PC SNOM images presented in Figure 5 was $\sim 100$ nm that coincides to the nominal aperture diameter $a$ of the SNOM probes used, which defines the spatial resolution of the method. Since the surface density of the smaller QDs was more than 2 times greater than the one of the bigger QDs, the images of QDs in Figure 5b tend to overlap. It is worth noting that not all QDs are manifested in the PC SNOM images, but only these ones, in which the ground state transition energy (or the one of another allowed interband optical transition between the quantum confined electron and hole states) matches the photoexcitation photon energy. The self-assembled InAs/GaAs(001) QDs are featured by a considerable natural scatter in size and, correspondingly, in the interband transition energies [9]. The width of the latter is much greater than the pumping LD emission linewidth ($\sim 10$ meV). As a result, only a small fraction of QDs falls into the LD emission line energy interval and are manifested in the PC SNOM images.

Figure 5. PC SNOM images (300 K) of the input window of a PD based on the $p$–$i$–$n$ structure with embedded InAs/GaAs(001) QDs. $\lambda_{ex}$, nm: (a) 1310; (b) 980.
The rest QDs are manifested in the PC SNOM image as the dark spots on more or less constant background (originating from the impurity PS of the $i$-GaAs layer and of the $n^+$-GaAs substrate as well as from the stray photoexcitation reflected back from the back side of the substrate) because of the scattering of the photoexcitation outgoing from the SNOM probe aperture on the individual InAs QDs [10]. This is a principal difference of the InAs QDs from the self assembled GeSi/Si(001) nanoislands studied in Ref. [3]. The latter are the type-II heterostructures, so that the initial hole states of the interband optical transitions are confined in the GeSi nanoislands, whereas the final states are the continuous ones in the conduction band of the host matrix material (Si). As a result, all GeSi nanoislands with the ground state interband transition energy $E_0$, being smaller than the photoexcitation photon energy, are manifested in the PC SNOM images whether the hole states in the GeSi nanoislands are the quantum confined ones or not [3]. In the relaxed InGaAs clusters, the size quantization effect is absent due to relatively large size of these ones as compared to the de Broglie wavelength of the electrons and holes. As a result, all relaxed InGaAs clusters are manifested in the PC SNOM images in Figure 5 as the white spots, which sizes and surface density ($\approx 4\times10^8$ cm$^{-2}$) coincide with the ones of the relaxed InGaAs clusters in the AFM image in Figure 2a.

4. Conclusion
The results of the present study demonstrate the possibility of observation of the photoresponse from single InAs/GaAs(001) QD due to the interband optical transitions between the quantum confined electron and hole states in the QD at the photoexcitation through a SNOM probe and of imaging the individual InAs/GaAs(001) QDs in the PC SNOM images. The spatial resolution of PC SNOM was limited by the SNOM probe aperture diameter $\sim 100$ nm. Not all QDs are manifested in the PC SNOM images but only these ones, in which the energy of the ground state interband optical transition (or another allowed interband transition) between the quantum confined electron and hole states match the photoexcitation photon energy. The rest QDs are manifested as the dark spots in the PC SNOM images due to the scattering of the light incident from the SNOM probe by the QDs.

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