Near-field magneto-photoluminescence of GaAs/AlGaAs/InP/GaInP$_2$ quantum well-quantum dot structures

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Abstract. This work presents the results of the growth, structural characterization and magneto-photoluminescence spectroscopy measurements of GaAs/AlGaAs/InP/GaInP$_2$ quantum well-quantum dot structures. We demonstrate that GaAs/AlGaAs QDs in these structures are formed above InP/GaInP$_2$ QDs. This allows us to measure the internal magnetic field in the InP/GaInP$_2$ QD by monitoring the Zeeman splitting of the excitonic transition in the GaAs/AlGaAs QD.

1. Introduction
Self-organized InP/GaInP$_2$ quantum dots (QDs) are natural anionic molecules (AMs) and are promising objects for research in the physics of strongly correlated electron systems and for the development of a topological quantum gate [1, 2]. AMs are formed in these QDs due to intrinsic electron doping and built-in magnetic field ($B_{0b}$) induced by atomic ordering in GaInP$_2$ [3]. In the photoluminescence (PL) spectra of InP/GaInP$_2$ QDs, the built-in magnetic field results in an anomalous shift of PL lines in a magnetic field, their circular polarization, and an anomalously small localization area in a zero magnetic field. It varies from 2 to 14 T, depending on the size of the QD and the number of electrons. In the present study, we report the development and characterization of a structure in which GaAs/AlGaAs QDs have been formed above InP QDs. It can be used as an external magnetometer of $B_{0b}$ by registration of the Zeeman splitting of the excitonic transition in GaAs QDs, for which such splitting is well known and reaches values up to 1.7 meV at 15 T [4, 5].

2. Experimental details
The “magnetometer” structure has been grown by metal organic vapor phase epitaxy (MOVPE) on a (001) GaAs substrate and consists of an InP/GaInP$_2$ QD layer, capped by a GaAs/AlGaAs quantum well (QW) (figure 1). The InP/GaInP$_2$ layer contains InP QDs formed by the deposition of 3 InP monolayers sandwiched between the bottom and top GaInP$_2$ claddings with a thickness of 500 and 40 nm,
respectively, lattice-matched to GaAs. The GaAs/AlGaAs part contains a 10-nm GaAs layer sandwiched between 20/40 nm Al$_{x}$Ga$_{1-x}$As ($x = 0.6$) claddings. Typically, the density of InP QDs is about $10^{8}$ cm$^{-2}$, and nearly 20% of them are AMs. AMs have a shape of a flat lens with a thickness of ~15 nm and a lateral size of ~150 nm with up to 10 electrons and 5-10 T $B_{0}$. We expected that QDs could be formed within GaAs QWs due to thickness fluctuations and strain inhomogeneity of the GaInP$_{2}$ layer.

Below we present the characterization of this structure obtained using transmission electron microscopy (TEM) and high-spatial-resolution PL.

Cross-sectional TEM samples were prepared by conventional mechanical polishing using a Model 590 Tripod Polisher followed by Ar$^{+}$ milling in a Gatan PIPS 691 precision ion polishing system. TEM specimens were characterized by a JEOL 2100F and a FEI G2 Talos microscopes operating at an accelerating voltage of 200 kV.

PL spectra were measured at ~10 K with a spatial resolution of 500 and 50 nm by means of a conventional micro-PL ($\mu$-PL) set-up and a near-field scanning optical microscope (NSOM), respectively. Micro-PL spectra were excited through a 50× micro-objective by a CUBE-405 (405 nm) semiconductor laser and recorded by a Trivista monochromator. NSOM spectra were measured in external magnetic fields (0-7 T) in the excitation-collection mode using fiber-optic probes, a CW Nd:YAG laser (532 nm) and a monochromator with a 500 mm focal length. The PL intensity map of a selected single QD was constructed with a step of 50 nm.

3. Structural data (TEM)

We can see from the cross-sectional TEM image (figure 1, lower right part) that the GaAs/AlGaAs QW exhibits a thickness variation on a length scale ~100 nm, which means that QDs are indeed formed in the QW layer. These GaAs/AlGaAs QDs are located in the vicinity of the InP QD. The thickness of the GaAs QD region is ~15 nm, while the surrounding “wetting layer” is about 5 nm. The TEM image also reveals an AlGaAs/GaInP$_{2}$ interface layer with a thickness of ~5 nm. The thickness of this layer also increases (up to 10 nm) above the InP QD and indicates QD formation. By analyzing the TEM energy dispersion spectra (not discussed here), we have found that the interface layer is a GaAsP one with a GaP composition of ~10-20%. The appearance of this layer can be explained by the memory effect in the MOVPE reactor after switching between the sources of group V atoms.

Figure 1. Arrangement of the layers and QDs (left) and cross-sectional TEM image (lower right) of the magnetometer structure. Upper right is the energy diagram of the conduction band of the AlGaAs QW, showing the QD formation in thicker parts.
4. $\mu$-PL spectra

Figure 2 presents a typical $\mu$-PL spectrum of our magnetometer sample. In the spectrum, we can see a set of bands (about ten) and sharp lines (about fifty) in the range of 650-950 nm. From our previous measurements of InP/GaInP$_2$ QD structures [1, 2], we can assign the lines in the range of 650-670 nm to the emission of localized excitons of GaInP$_2$, and in the range of 670-750 nm to InP/GaInP$_2$ QDs. We also found in preliminary PL measurements of GaInP$_2$/AlGaAs structures (i.e. the structures with only heterointerface, not discussed here) that the spectral range of spontaneous emission of the GaAsP heterointerface layer overlaps with the spectral range of InP/GaInP$_2$ QDs and, thus, some fraction of the sharp lines observed in the 700-750 nm region can be assigned to these QDs. A similar result of the GaInP$_2$/AlGaAs interface study was also reported previously [6].

![Figure 2](image_url)

**Figure 2.** The $\mu$-PL spectrum of a QD-QW structure at 10 K. The insert shows the spectral region containing a possible Zeeman doublet.

The features related to the magnetometer are the lines in the 750-810 nm range, which, together with the band at 820 nm, can be assigned to the GaAs/AlGaAs QDs and QW, respectively. We have also observed a series of peaks at 830-900 nm, which, due to their longer emission wavelength, can be assigned to the GaInAs QDs, formed possibly due to As diffusion into the GaInP layer or residual agglomeration of In atoms on the GaInP free surface.

In the region of GaAs/AlGaAs QDs, we found several doublets with an energy splitting of ~1 nm (insert in figure 2), which can be a result of the Zeeman splitting induced by AMs $B_{\text{bs}}$. 
5. NSOM magneto-PL spectra

In the NSOM PL spectra (figure 3), the number of lines and peaks is reduced (several times) due to the higher spatial resolution. In the spectra shown in figure 3, the lines related to GaInP$_2$ are not resolved, and the peak of the GaAs QW has a very week intensity compared to the $\mu$-PL spectra, which is caused by the lateral inhomogeneity of the structure. Due to this inhomogeneity, we did not find doublets in NSOM PL in the range scanned (1.5×1.5 $\mu$m$^2$) (see below).

![Graphs showing NSOM magneto-PL spectra](image)

**Figure 3.** (a) PL NSOM spectra at 10K for two different tip locations. (b) PL spectra of the single GaAs QD at magnetic fields of 0, 1, … and 7 T. (c) Magnetic field dependencies of the spectral positions of the Zeeman splitting components $C_L$ and $C_H$. Notation $C_{ds}$ corresponds to the value of $(C_L + C_H)/2$. 


However, we have measured the dependence of the PL line of a single GaAs QD in an external magnetic field in the range 0-7 T (see inserts in figure 3, b), which confirms their formation. We have observed a blue (diamagnetic) shift and the Zeeman splitting of the line of a single GaAs QD at a magnetic field above 4 T. The shift is quadratic and has value $\beta = 9 \mu eV / T^2$.

The diamagnetic shift depends on the exciton wave function $\langle x^2 \rangle$, namely, $
abla = e^2 \langle x^2 \rangle / 8 \mu$, where $e$ is the charge of the electron and $\mu$ is the reduced mass of the exciton. The exciton radius is given by $r = 2\sqrt{\langle x^2 \rangle}$ [7], and, assuming the exciton reduced mass in GaAs equal to $m_e/3.45$, we can estimate the exciton lateral size at about 90 nm, which agrees well with the TEM data (see figure 1).

6. NSOM imaging

Figure 4 presents the NSOM PL intensity maps of the lines of single InP, GaAs and GaAsP QDs emitting at 730, 778 and 737 nm, respectively, measured in the area $1.5 \times 1.5 \mu m^2$. It is seen that all the QDs are located at the upper edge of the map and have a size of 30, 300 and 100 nm, respectively, while the GaAs and GaAsP dots are separated from the InP QD by ~1000 and ~100 nm, respectively. This means that the GaAsP QD is formed closer to the InP one, which is in coincidence with the TEM data.

The measured InP QD has an anomalously large size, which indicates non-optimal growth conditions that suppress the formation of AMs and neighboring GaAs QDs. This highlights the importance of the optimization of the growth conditions toward uniformity improvement.

The data in figure 4 demonstrate the unique capabilities of the NSOM technique in characterization of the magnetometer structure.

![Figure 4. PL NSOM intensity maps (size 1.5×1.5 μm²) of three single dots lines having emission energy of 778, 730 and 737 nm.](image)

7. Conclusion

The GaAs/AlGaAs/InP/GaInP₂ quantum well-quantum dot sample was grown and the method of using a GaAs QD as a magnetometer of the built-in magnetic field in InP QDs was tested. The formation of GaAs QDs and their Zeeman splitting was demonstrated. It has been shown that the position of a single QD, its relative location and dimensions can be determined with high precision using NSOM.
Acknowledgments
This study was supported by the Russian Science Foundation, project no. 19-19-00246. The $\mu$-PL measurements were performed in terms of the Russian Foundation for Basic Research, project no. 18-02-01212.

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