Natural quantum dots in the InAs/GaAs wetting layer

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We report on microphotoluminescence study of excitons localized by potential fluctuations in a wetting layer (WL), which accompanies InAs/GaAs quantum dots (QDs). Linear polarization of spectral lines due to localized excitons enable us to identify a neutral excitonic and biexcitonic emission. A charged exciton has also been identified. High resolution transmission electron microscopy measurements of the investigated structure reveal lateral fluctuations of In content in the WL, as well as its broadening. Both effects give rise to potential fluctuations, which can confine excitons observed in our measurements. The potential fluctuations can be regarded as "natural" QDs in the WL. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2918836]

Semiconductor self-assembled quantum dots (QDs) have been a hot topic of research over the last decade.[1] This results from interesting fundamental physics and numerous potential applications. The most intensely investigated system comprises of InAs QDs grown in GaAs matrix. Spontaneous formation of InAs/GaAs QDs in a Stranski–Krastanow growth mode[2] takes place after the deposition of a thin InAs/GaAs layer, referred to as the wetting layer (WL). Despite its important effect on electrical and optical properties of self-assembled QDs,[3] considerably fewer results are available on the electronic structure of the WL. One of the factors, which may potentially have important implications for properties of self-assembled QDs, is a three-dimensional localization of excitons in the WL, which is equivalent to the formation of "natural" QDs (NQDs)—as opposite to self-assembled QDs. Some indications of three-dimensional localization effects in a WL have previously been observed in the InAlAs/AlGaAs (Ref. 4) and the InAs/GaAs (Ref. 5) structures. The possible formation of a charged exciton in the InAs/GaAs NQD was also proposed.[6] In this letter, we report on spectroscopic studies of excitons localized in the WL. We identify a neutral exciton and biexciton, as well as charged exciton emission lines. We also present results of high resolution transmission electron microscopy (HRTEM) analysis, which confirms In-composition fluctuations in the WL. These composition fluctuations, in our opinion, are responsible for the potential fluctuations that give rise to the formation of NQDs in the WL.

The sample investigated in this work was grown by using molecular beam epitaxy.[7] It contains a single layer of InAs QDs grown at 524 °C on a n^+ GaAs substrate, 800 nm GaAs buffer layer, and capped with a 100 nm GaAs top layer. In flush has been applied to the QDs at 5 nm. A set of mesa structures were prepared on the sample in order to reproducibly locate a dot, optically addressed. The He–Cd laser ($\lambda=442$ nm) was used for photoluminescence (PL) excitation. The PL was measured at $T=6$ K. The excitation light was delivered and the PL was collected via a microscope objective (spot size ~ 1 μm), dispersed by using a 0.5 m monochromator and detected by using a charged coupled device (CCD) camera. The polarization of emitted light was analyzed by using a linear polarizer placed before the spectrometer.

The PL spectrum from the investigated structure consists of the emission from the GaAs barrier, self-assembled QDs,[8] and the emission related to the WL. The typical PL spectrum in the energy range of the expected WL emission (1.42–1.44 eV) measured on a submicron-size mesa (600×600 nm^2) consists of several sharp lines [see Fig. 1(a)]. We relate the features to the recombination of electrons and heavy holes localized in the InAs/GaAs WL.[9] The discrete structure of the emission suggests that the WL is strongly disordered. The disorder leads to potential fluctuations, which localize excitons, as previously observed in disordered thin GaAs/AlGaAs quantum wells.[10] The spectrum measured on a larger mesa (10×10 μm^2) evolves into the quasicontinuum of a broad peak [see Fig. 1(b)]. Moreover, at some spots on the investigated mesas, several well-resolved lines emerge in the spectrum at lower (up to 10 meV) energies. Emission lines from the low-energy tail of the spectrum must be due to the recombination of excitons in the deepest potential fluctuations. Their separation from the main emis-

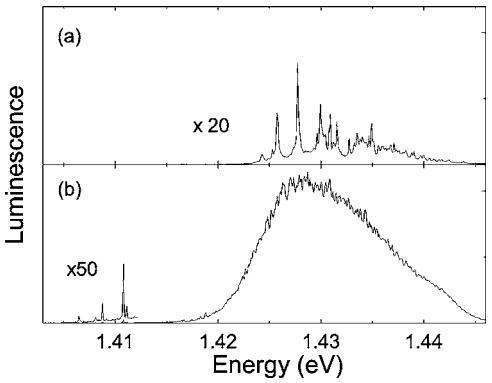

FIG. 1. Micro-PL spectrum ($T=6$ K) of a structure with self-assembled InAs/GaAs QDs in the WL energy range. (a) The spectrum from the 600×600 nm^2 mesa ($I=320$ W/cm^2). (b) The spectrum from the 10×10 μm^2 mesa ($I=240$ W/cm^2).

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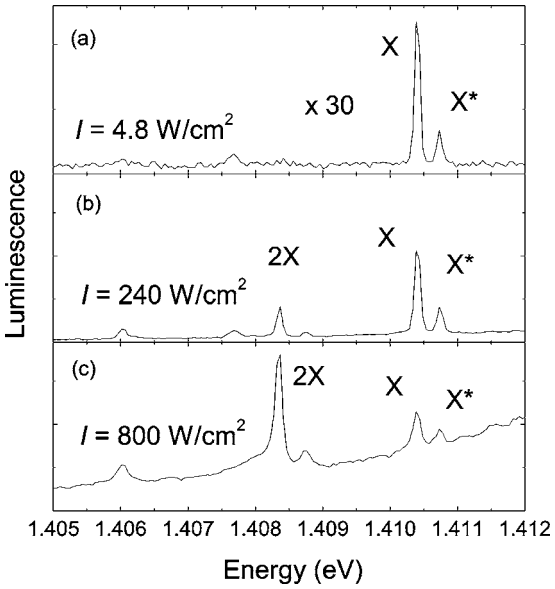

FIG. 2. Micro-PL spectra due to the excitons localized in a single NQD in the InAs/GaAs WL at $T=6$ K excited with (a) 4.8, (b) 240, and (c) 800 W/cm^2 excitation power density. The spectrum at low excitation power is dominated by neutral (X) and charged (X^*) exciton lines. At higher power density, a neutral biexciton ($2X$) emerges, which dominates the spectrum at the strongest excitation.

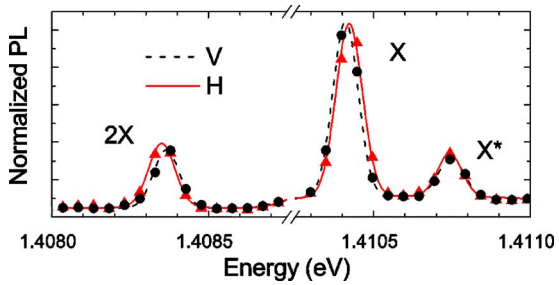

FIG. 3. (Color online) Normalized polarization-sensitive micro-PL spectra of the excitons localized in a single NQD in the InAs/GaAs WL. The circles and triangles correspond to experimental points for the vertical and horizontal linear polarizations of detected light. The dotted and full lines are Gaussian fits of the corresponding spectra.

sion peak enables more detailed analysis of their properties.

The excitation power dependence of the spectrum due to the localized excitons is shown in Fig. 2. At low excitation power density [4.8 W/cm^2, see Fig. 2(a)], the spectrum is dominated by two emission lines: X and X^*. The inspection of the CCD image confirmed that both lines originate from the same spatial position of the structure. Similar structure of the PL, comprising of two lines, has been found on several spots of the sample. These observations confirm that the two features are due to the recombination of excitons in the same object. Both lines gain in intensity with increasing power [see Fig. 2(b)]. Simultaneously, another emission line ($2X$) emerges at lower energy (separated from X by -2.0 ± 0.1 meV). The $2X$ emission line dominates the spectrum excited with the highest power density (800 W/cm^2) [see Fig. 2(c)]. To establish the exciton occupancy for the X, X^*, and $2X$ emission lines, more detailed power-dependent measurements have been performed. Intensities of the X and X^* lines sublinearly increase (exponents of a power law $n_X=0.89$ and $n_{X^*}=0.83$) before simultaneously reaching a maximum and decreasing for higher excitation density. This suggests that both lines arise from a single exciton occupancy of the dot. On the contrary, the $2X$ line intensity superlinearly increases (the exponent $n_{2X}=1.55$), which confirms its attribution to the biexciton recombination.

In order to identify the charge state of observed excitons, we have performed polarization-sensitive PL measurements. It has been observed that the X and $2X$ emission line are composed of two components, which are linearly polarized in perpendicular directions [see Fig. 3]. The Gaussian fits of the components give the value of the splitting equal to 15 ± 5 μeV. The signs of the X and $2X$ emission lines splitting are opposite. The splitting must result from anisotropic exchange in electron-hole interaction, which is a characteristic for neutral excitons in QDs.[11] The splitting pattern allows us to attribute the X and $2X$ emission lines to the neutral exciton and biexciton recombination in a QD. It has been found that within experimental accuracy, the optically-distinguished directions of the sample correspond to the [110] and [1-10] crystallographic axes of the sample. This confirm expected elongation of the localization potential wells along one of those axes due to anisotropic diffusion coefficient of In on the (100) GaAs surface.[12] The optical asymmetry has not been observed in the case of the X^* emission line. This suggests its attribution to a trion (charged exciton) recombination in the QD. Observation of both the neutral and the charged excitons is usually reported in spectroscopic studies of nonresonantly excited QDs (Ref. 13) and the relatively low exchange-interaction splitting of a neutral exciton is similar to the values found in NQDs formed by interface fluctuations in thin quantum wells.[11]

We relate the localization of excitons to potential fluctuations in the investigated InAs/GaAs WL. To elucidate the source of those fluctuations, we have performed the HRTEM analysis of the structure. Typical cross-section HRTEM image in the [110] zone axis of the WL is presented in Fig. 4(a). Local lattice distortion measurements were performed by the peak-finding procedure similar to that described in Ref. 14. The In composition was recalculated from the distortion assuming that Vegard's law and the intermedial stress-relaxation model are valid, which seems to be appropriate for a relatively thick sample that has been used in our 300 kV microscope. The color-coded map of the indium fraction x corresponding to the HRTEM image from Fig. 4(a) is shown in Fig. 4(b). The highest indium fraction in the WL (shown in red online) reaches the level of 0.35–0.40. The regions are about 2–3 nm large and 1 nm high. The indium fraction in other areas of the WL (shown in green online) reaches 0.2. The calculated standard deviation of indium fraction in the frames A and C (in the GaAs barrier) are equal to 0.049 and 0.048, respectively, whereas, frame B inside WL is considerably larger and is equal to 0.066. Therefore, it is reasonable to consider that observed indium fluctuations in the WL are larger than the image noise observed in the GaAs areas. The averaged profile [which corresponds to the HRTEM image from Fig. 4(a)] of the indium fraction x as a function of position in the growth direction is presented in Fig. 4(c). The asymmetric shape of the profile can be discerned and is due to segregation of indium during the growth. The profile was characterized by using phenomenological Muraki model of indium segregation.[15] As a result, the nominal indium concentration $x_0=0.374$, the WL width 2.9 ML, and segregation coefficient $R=0.716$ have been obtained. The segregation coefficient corresponds to the value reported in Ref. 16 for a

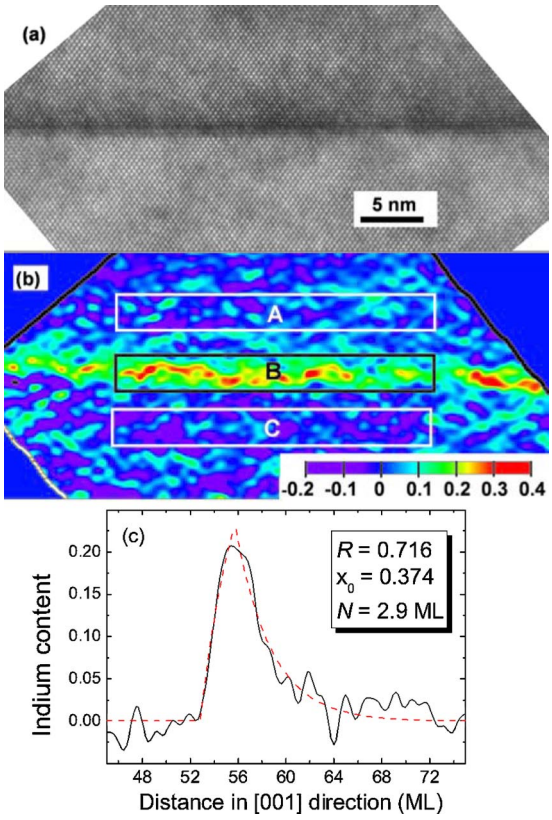

FIG. 4. (Color online) The HRTEM analysis of the InAs/GaAs WL. (a) The HRTEM image of a typical region of the WL. (b) The corresponding map of the indium fraction x, obtained by the quantitative image analysis. (c) The indium composition profile of the WL obtained by laterally averaging the map from the one presented in (b) (continuous line) and the corresponding Muraki fit (dashed line).

structure grown in similar conditions. The integrated total indium content of the WL that was deduced from our analysis corresponds to 1 ML. On the other hand, based on the amount of In deposited, and its expected volume transferred, to QDs, the amount of In should be closer to 1.4–1.6 ML. This discrepancy may be due to the proximity of the inspected area to QDs, which captured more In from this area than expected in average. Another reason could be a limited validity of the assumptions on elasticity used for our analysis, image distortion, additional relaxation at the sample surface, etc.[17] Importantly, this quantitative discrepancy does not impact our main observations, which are the WL broadening and the existence of lateral fluctuations of its In composition. Both the broadening and the lateral fluctuations of In concentration in the WL have been previously reported.[16,18] These fluctuations affect the band structure by composition and strain effects, and give rise to three-dimensional localization of excitons in the WL. The localization is equivalent to the formation of NQDs and we believe that their presence should be taken into account while analyzing the properties of self-assembled QDs.

In conclusion, we have performed micro-PL measurements on the structure with self-assembled InAs/GaAs QDs. We have found the excitonic emission, which has been ascribed to NQDs as the InAs/GaAs WL. We have identified the neutral exciton and biexciton, as well as a trion. It has been proposed that potential fluctuations in the WL are responsible for the formation of the NQDs. The HRTEM analysis confirmed the presence of broadening and lateral composition fluctuations for the WL.

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