

## Pauli-blocking imaging of single strain-induced semiconductor quantum dots

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The photoluminescence (PL) of InP strained-induced quantum dots in a GaInAs/GaAs quantum well is measured at low temperature (4.2 K) using near-field scanning optical microscopy. The PL originating from the first three confined levels of eight individual dots is mapped out over an area of  $1.4 \times 1.4 \mu\text{m}$ . The spatial resolution of the PL of the lowest energy level is found to be limited to about  $0.5 \mu\text{m}$ . In contrast, the mapping of the PL of the higher excited state shows a much improved spatial resolution of the order of 150 nm which is the instrument resolution. This effect is understood in terms of Pauli-blocking of the dot level filling. © 1999 American Institute of Physics. [S0003-6951(99)00721-4]

Photoluminescence (PL) is a characteristic optical signature of interband electron-hole exciton energy in semiconductor heterostructures. Typical PL spectra contain information related to the local band structure, the Fermi energy and spatial confinement at the position where the electron-hole recombine radiatively. Consequently, high resolution spatial mapping of the PL can be used as a powerful characterization technique of semiconductor heterostructures.<sup>1</sup> In particular, there is at the present time much to be learned from microscopic investigations of the electronic properties of self-assembled semiconductor quantum dots.<sup>2</sup> High resolution PL microscopy can be achieved using a near-field scanning optical microscope (NSOM) with optical probes illuminating locally the sample (illumination NSOM) with aperture sizes ranging between 100 and 200 nm.<sup>1</sup> The spatial resolution with which NSOM PL studies can be conducted in semiconductors is usually ultimately limited not by the size of the optical probe, but rather by the diffusion lengths of the photogenerated carriers.<sup>1</sup> Typical diffusion lengths of excitons can range from a few hundreds of nanometers to several micrometers. The spatial information that can be retrieved from PL microscopy is therefore nonlocal within this diffusion length. The problem of diffusion limited spatial resolution of the PL mapping can be circumvented by collecting the PL through the tip aperture<sup>3</sup> (collection NSOM) but usually with a very poor collection efficiency.

In this work we report on a case of local illumination PL spectroscopy in which the limitation of spatial resolution due to exciton diffusion can be relaxed by making use of Pauli-blocking of the level filling in semiconductor quantum dots.

In such dots, electrons and holes are quantized in discrete energy levels.<sup>4</sup> Like in atoms, the occupation of each energy level is governed by the Pauli exclusion principle so that when a given level is full, electrons or holes have to fill the next lowest unoccupied available energy state. Manifestation of this Pauli-blocking of levels can be directly measured in PL<sup>5</sup> or interband-absorption spectroscopy.<sup>6</sup> Ideally, the lowest energy level of a dot located within exciton diffusion distance from the probe contribute invariably to the PL. In contrast, the contribution of the higher energy dot levels depends on the number of electrons and holes filling of the ground state which is an exponential function of the probe-dot distance along the sample surface. This strong nonlinear response of the higher energy dot levels PL to the tip-dot distance is used here to improve the PL spatial resolution beyond the diffusion length limit.

The sample under investigation is shown in Fig. 1. It consists of self-assembled InP dots grown on a  $\text{Ga}_{0.1}\text{In}_{0.9}\text{As}$  well sandwiched between GaAs barriers.<sup>7</sup> The photogenerated electrons and holes are confined along the growth axis in the 6.5-nm-thick  $\text{Ga}_{0.1}\text{In}_{0.9}\text{As}$  layer. The dot lateral confinement is achieved within the GaInAs layer directly under the InP dots by the strain field originating from the lattice mismatch between the InP islands and the  $\text{GaAs}/\text{Ga}_{0.1}\text{In}_{0.9}\text{As}$  layered structure.<sup>7</sup> The typical density of the InP islands is  $1.0 \times 10^9/\text{cm}^2$  making the dot-dot average separation about 350 nm. Far-field measurements show that up to five dot confined levels can contribute to the PL under appropriate laser excitation power.<sup>7</sup>

The PL measurements were performed using a top-loading low-temperature NSOM. The microscope fits inside a 50.8 mm outside diameter vacuum tight insert which is plunged in a liquid He bath cryostat. The sample and the microscope are cooled down to 4.2 K in He exchange gas.

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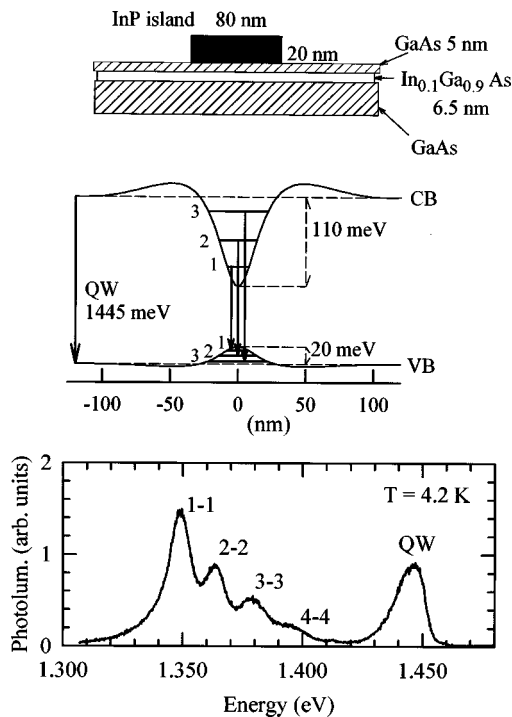


FIG. 1. Top: Schematic representation of an InP stressor island on a GaInAs/GaAs two-dimensional quantum well. Middle: The corresponding schematic band diagram is shown in the plane of the InGaAs just under the InP island. The measured PL energies are represented by the arrows. Bottom: typical 4.2 K far-field PL data.

High spatial resolution is achieved by bringing the aperture at the apex of an aluminum coated tapered optical fiber tip<sup>8</sup> into a close proximity with the sample surface.<sup>8</sup> The tip-sample distance control is achieved using a piezoelectric tuning fork force sensor<sup>9</sup> allowing to measure the tip-sample friction and shear forces ranging from about 0.1 pN to several nanonewtons. The tuning fork holding the tapered optical fiber tip is directly mounted on a specially developed low temperature preamplifier in order to minimize the parasitic capacitive cable losses of the weak piezoelectric shear and friction force signal. In a typical friction force imaging mode, the tip aperture peak to peak excursion at the resonance frequency of the fork (33 kHz) is adjusted with a mechanical dither excitation to be 50 pm. The friction force is kept at 30 pN using a feedback loop on the tip-sample distance. It is worth mentioning that forces exceeding 1 nN lead to an irreversible modification of the InP dots. The optical probe was used in illumination mode as to excite locally the sample surface with the 632.8 nm line of a HeNe laser. The size of the tip aperture was  $160 \text{ nm} \pm 10 \text{ nm}$  as characterized from far-field goniometrical measurements.<sup>10</sup> We purposely used a relatively large aperture in order to get enough pumping power to populate at least the three lowest quantized levels when the tip was positioned over a InP island. The total power was measured to be 50 nW corresponding to about  $250 \text{ W/cm}^2$ . The reflected signal containing the PL signal was collected over 70% of the total solid angle available in reflection using an ellipsoidal aluminum mirror. The PL signal was then dispersed on the grating of a 640 mm focal length spectrometer of numerical aperture 0.1, and detected with a nitrogen cooled front-illuminated Si-chip charged coupled device camera. The spectrometer resolution was set

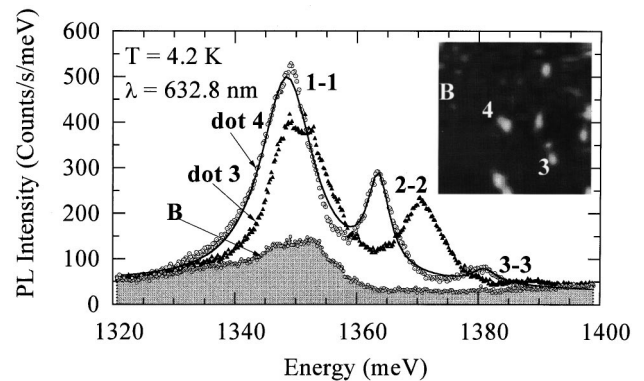


FIG. 2. The inset shows a topography of the sample measured at 4.2 K over a field of  $1.4 \times 1.4 \mu\text{m}$ . The white areas are the InP stressor island protruding 20 nm above the substrate area (black). The apparent elongated shape of the InP island is due to the NSOM-tip. The PL spectra measured on two different dots and on a dot-free region (B) are shown. The full line is a fit with three Lorentz shaped peaks.

to 0.5 meV. The sample was mounted on a piezoelectric scanner providing a total of  $1.4 \mu\text{m} \times 1.4 \mu\text{m}$  scan range from under 0 to +100 V drive signal. Rough positioning of the sample was achieved over  $5 \times 5 \times 5 \text{ mm}$  in  $x$ ,  $y$ , and  $z$  using a low-temperature, low-voltage piezoelectric inertial positioner. The PL signal was integrated over 10 s/tip position-pixel over the whole measured spectral range. The PL images shown in this work were made over a range of  $20 \times 20$  pixels with  $70 \text{ nm} \times 70 \text{ nm}$  size.

The inset of Fig. 2 shows a constant friction-force image of the sample surface which is assumed to render the topography. There are in this field eight InP dots protruding 20–25 nm with a diameter of about 200 nm. The measured dot heights are consistent with high resolution noncontact scanning topographies. The measured average lateral size of the dot is larger than the actual dot size of 80 nm due to the tip size at its apex. The luminescence spectra corresponding to three extreme situations within the measured field of dots are shown in Fig. 2. In the position marked (3) the measured luminescence shows three distinct peaks that are interpreted as resulting from the interband radiative recombination of the dot excitons in the lowest three energy level in accordance with far-field measurements.<sup>5</sup> The observation of the 2–2 and 3–3 transition is to be understood in terms of Pauli-blocking of the level population. In the position marked (4) the PL spectrum of a different dot is shown. Although the position of the 1–1 peak seems to be almost unchanged, the 2–2 and 3–3 peaks are about 7 meV blueshifted. At the location marked (B) there is no InP island as seen from the topography. At this tip position, the PL corresponding to the 2–2 and 3–3 transition is no longer present while the 1–1 transition, although weaker, is still there. This makes it clear that the photogenerated excitons can diffuse during their lifetime from the position from where they were excited. The disappearance of the 2–2 and 3–3 PL peaks at position (B) shows also that the rate at which the carriers reach the nearest dots is low enough so that no Pauli-blocking occurs. The exciton diffusion radius is what limits the spatial resolution of the PL imaging of the ground state to the diffusion length. This limitation in the spatial resolution is well rendered in the Fig. 3(a) where the integrated intensity of the ground state PL is imaged as a function of the tip position. From

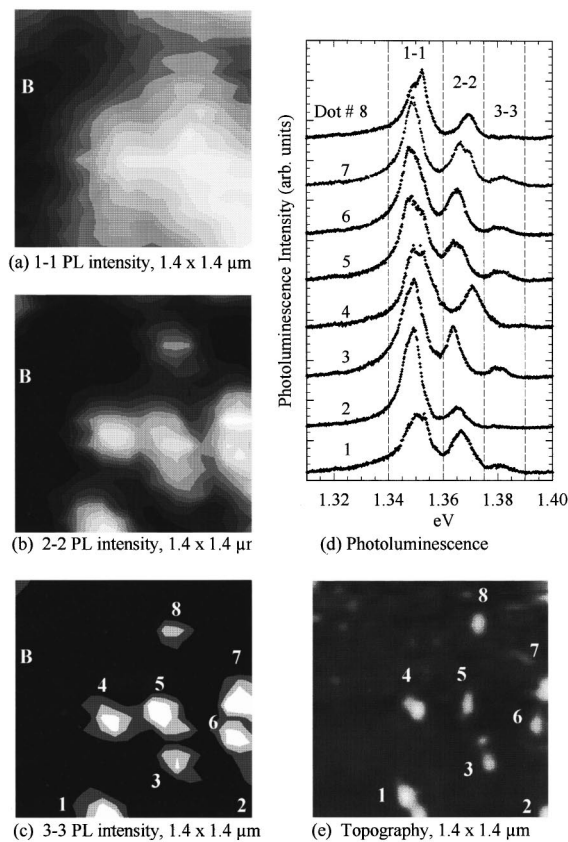


FIG. 3. (a), (b), and (c) are the integrated PL intensity maps of the 1–1, 2–2, and 3–3 emission peaks, respectively. The integration range is shown in (d) between the vertical dashed lines. The PL spectra corresponding to the eight dots position marked in (c) are shown in (d). The sample topography shown in (e) is fully correlated with the PL map of (c).

such an image the diffusion length radius is estimated to be around  $600 \text{ nm} \pm 100 \text{ nm}$ . In contrast to this, the maxima in the PL intensity of the 3–3 transition in Fig. 3(c) and the topographical signature of the InP dots are fully correlated. In this case the spatial resolution is limited by the size of the tip aperture. A clear trend in this correlation persists when looking at the 2–2 transition. Clearly, the spatial resolution in PL microscopy of quantum dots is greatly improved by Pauli-blocking effects. The mapping of the PL of the higher excited states demonstrates that the 2–2 and 3–3 PL lines in the spectra of Figs. 3(b) and 3(c) are largely due to single quantum dots.

In contrast to recent PL measurements<sup>4</sup> performed on single semiconductor quantum dots which show PL line widths well below  $100 \mu\text{eV}$ , we measured here PL full widths at half maxima of 10, 4.5, and 5 meV for the 1–1,

2–2, and 3–3 PL peaks, respectively. In the far-field line widths were found to be 13, 9, and 13 meV, respectively. The broad PL line width of an ensemble of dots is usually understood in terms of inhomogeneous broadening due to spatial fluctuations of the dot potential. Such statistical inhomogeneous broadening leads usually to Gaussian shaped PL peaks. As opposed to this, lifetime level width broadening of the PL is expected to produce Lorentz shaped emission peaks. The PL lines of Fig. 2 are fitted with such a Lorentz shape. By comparison, Gauss line shapes are found to produce a much poorer fit within our PL noise limit. This suggests that the broadening is not due to spatial nor temporal fluctuations slower than the scale of the exciton life time ( $\sim 1 \text{ ns}$ ). At this point we have no definite explanation for the origin of such a broadening. It is worth pointing out that in the work of Hess and coworkers<sup>1</sup> a similar type of broadening is observed for “natural” quantum dots in GaAs/AlGaAs quantum well located within 5 nm from the sample surface. In their work, dots located deeper under the surface shows much narrower PL line. We speculate that in our samples, the potential experienced by the dot is locally disturbed by near surface impurity states which charge fluctuates on time scales faster than the exciton radiative lifetime. The charge fluctuations of near surface impurities would be photogenerated by the probe itself at a rate not exceeding that of the number of impinging photons (i.e.,  $50 \text{ ps}^{-1}$ ). This, however, needs to be verified by performing timed resolved PL measurements on single quantum dots.

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