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Photoluminescence from a single InGaAs epitaxial quantum rod

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Microphotoluminescence (μ-PL) experiment has been performed on a structure with InGaAs/GaAs epitaxial quantum rods (quantum dots with the aspect ratio as high as 4.1) grown by depositing short-period InAs/GaAs superlattice by molecular beam epitaxy on GaAs substrate. The exciton and biexciton emission from a single quantum rod has been detected via the excitation power dependence of the μ-PL spectra. The origin of the single rod lines has been confirmed by a rate equation model. For a number of quantum rods within the investigated ensemble, the biexciton binding energy has been determined to be in the range of 1.0–2.2 meV. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2832635]

The three dimensional character of carrier confinement together with the ensemble natural inhomogeneities attracted much attention on self-assembled quantum dots (QDs), resulting in many application possibilities in modern electronics and optoelectronics (e.g., in lasers for fiber-based telecommunication and data transmission). Single QDs are also an excellent testbed for the investigation and control of solid-state quantum bits and for quantum information processing. The diameter and height of Stranski–Krastanow (SK) QDs can only be partially controlled by the growth conditions changes, however, their aspect ratio is mostly limited to well below 1. A technique of cycled deposition of a short-period InAs/GaAs superlattice on top of a seed SK QD layer[1,2] provides a tool for the independent control of the QD height and its diameter. In this case, the preferential incorporation of In adatoms in strained areas on top of the seed QDs leads to the formation of "column-shaped" nanostructures. Recently, this technique was extended to demonstrate InGaAs QDs with an aspect ratio exceeding 1,[3–6] leading to the formation of vertical quantum rods, also called "quantum posts." Additionally, it was shown that the In content profile in the vertical direction can be controlled by varying the deposition duty cycle.[6] This opens the way to the artificial engineering of the potential in the vertical direction and, thus, to the control of electron and hole wavefunctions, e.g., for the oscillator strength optimization, Stark shift, etc. This opportunity could be particularly relevant in the field of the coherent control of charge and spin states in single QDs. Hereby, there is presented a report on the investigation of exciton (*X*) and biexciton (*XX*) emission in single quantum rods (QRs) with an aspect ratio as large as 4.1. The experimental observations are supported by modeling of the *X*-*XX* dynamics, and prove the zero-dimensional character of electronic states in epitaxial QRs.

The investigated quantum rods have been grown by solid source molecular beam epitaxy on the (001) oriented GaAs substrate. The seeding InAs self-assembled quantum dot layer was deposited on a GaAs buffer and then followed by a sequence of 0.62 ML of InAs and 3 ML of GaAs repeated 35 times in order to obtain high aspect ratio object. More details of the growth process have been already reported elsewhere.[5,6] Transmission electron microscopy (TEM) has been performed in order to determine the rods' geometry and composition. The plane-view images revealed that they have a diamond (rhombus) shape with diagonals along the [110] and [1-10] directions and their length ratio is typically 1.22.[5] The cross-sectional TEM images (see Fig. 1) show they have the aspect ratio larger than 4 (height of 41 nm and the diagonal length of 10 nm, approximately) and an almost constant In content in the quantum rod (~40%) and in the surrounding two-dimensional (2D) "immersion" layer (~15%).[6]

Microphotoluminescence measurements (μ-PL) have been performed on an ensemble of quantum rods using a high numerical aperture and a long working distance microscope objective to focus the excitation beam (660 nm line of a semiconductor laser) and to collect the luminescence from the structure. The spot size is diffraction limited which gives the actual diameter of about 2–3 μm on the sample surface. An InGaAs liquid nitrogen cooled linear charge-coupled device has been used as a detector, which combined with a 0.55 m focal length single-grating monochromator allowed a spectral resolution below 100 μeV (typically enough for single quantum dot spectroscopy). The sample has been mounted in a microscopy type cryostat for low-temperature and short-focal length optical experiments. The setup is additionally equipped with a special antivibration and antidrift system providing long time stable position of the excitation beam on the sample surface.

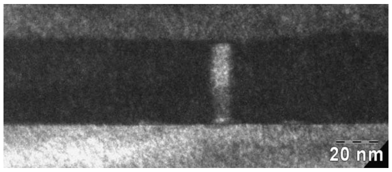

FIG. 1. (a) Cross-sectional TEM image of a single quantum rod grown by submonolayer close stacking of InAs and GaAs (along the [110] direction).

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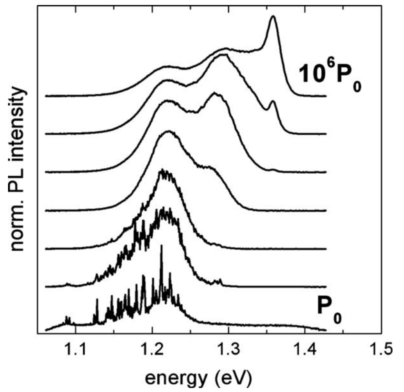

FIG. 2. Low temperature (5 K) μ-PL spectra of an ensemble of quantum rods in the range of the ground state emission for different values of excitation power.

Figure 2 shows a set of low temperature (5 K) μ-PL spectra taken as a function of the excitation power in a low spectral resolution mode (wide spectral range). For very low excitation powers, the spectrum splits into a number of single lines (still a few hundreds of rods can be illuminated within the spot diameter). The increase of the excitation power results in the appearance of the emission from higher-energy states and from multiparticle exciton complexes, and also in the enlarged effectively illuminated area and, therefore, in the increased total number of emitting rods. The QR ensemble spectrum exhibits a Gaussian-like peak centered at 1.22 eV for the intermediate excitation powers and the appearance of two additional peaks on the high energy side with further excitation increase. Based on the previous photoreflectance measurements and the energy level calculations of this kind of structures,[7] the peak around 1.285 eV can be attributed to the emission from the excited states within the QRs, whereas the peak at 1.36 eV to the emission from the ground state transition in the 2D layer surrounding the dots (forming a regular quantum well).

In order to perform single quantum dot study without sample patterning, it was necessary to focus on the low energy tail of the main QR emission peak where the average number of emitting rods is relatively small and PL lines related to excited states are not expected. Figure 3 shows a set of such spectra recorded as a function of the excitation power (100 nW to above 1 μW measured outside the cryostat). A typical exciton (X) and biexciton (XX) emission behavior is observed for these two lines (no other lines are detected within several meV range).

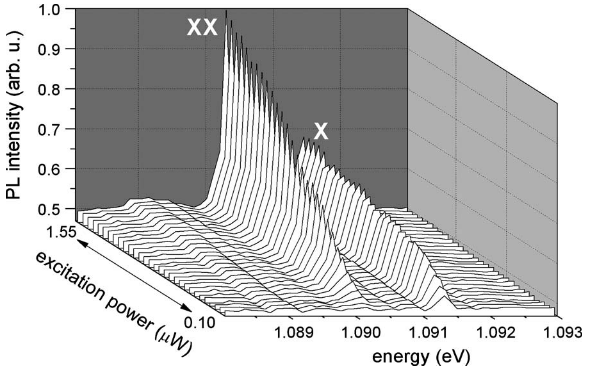

FIG. 3. Microphotoluminescence spectra as a function of the excitation power revealing emission lines corresponding to exciton and biexciton radiative recombinations in a single quantum rod.

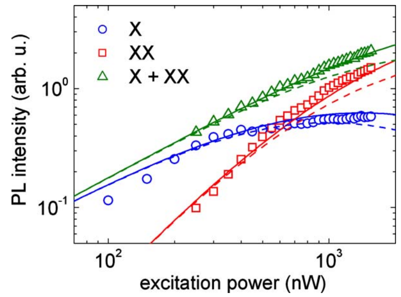

FIG. 4. (Color online) Integrated intensity dependence of both X and XX lines of a single quantum rod observed in the μ-PL spectra (symbols) and calculated according to the Eqs. (3) and (4) for $\tau_X/\tau_{XX}=2$ (dashed lines) and $\tau_X/\tau_{XX}=4$ (continuous lines).

In order to make a more quantitative analysis, the PL intensity dependence versus the excitation power has been plotted in Fig. 4. The intensity of the X line, which is attributed to the exciton radiative recombination, increases with power approximately linearly up to a power of about 350 nW, above which it saturates to an almost constant level. The intensity of the XX line follows a quadratic dependence up to 600 nW and also reaches the saturation range (increasing further but with a smaller slope). Additionally, a sum of both intensities ($X+XX$) has been plotted and exhibits almost linear behavior in the entire range of excitation. In order to support the interpretation and confirm if both the lines can be indeed related to the exciton and biexciton in the same quantum rod, the intensity versus excitation power dependence has been modeled using the following rate equations for an exciton:

$$\frac{dp_X}{dt} = -\frac{p_X}{\tau_X} + \frac{p_{XX}}{\tau_{XX}} + gp_0 - gp_X \tag{1}$$

and a biexciton,

$$\frac{dp_{XX}}{dt} = -\frac{p_{XX}}{\tau_{XX}} + gp_X, \tag{2}$$

where p_0 is the probability of the system being in its ground state (no exciton in the dot), $p_{X(XX)}$ is the probability of the dot occupation with an exciton (biexciton), $\tau_{X(XX)}$ is the respective radiative lifetime, and g is the generation rate (proportional to the excitation power). The emission intensity is related to the occupation probability and the state lifetime by $I_{X(XX)} \propto p_{X(XX)}/\tau_{X(XX)}$. For the stationary conditions and neglecting the probability of the higher levels occupation (which means $p_0+p_X+p_{XX}=1$) the following relations for the intensity of exciton and biexciton emission can be obtained:

$$I_X \propto \frac{g}{1+g\tau_X+g^2\tau_X\tau_{XX}}, \tag{3}$$

$$I_{XX} \propto \frac{g^2\tau_X}{1+g\tau_X+g^2\tau_X\tau_{XX}}, \tag{4}$$

which in the range of low generation rates reduce to the linear and parabolic dependencies, respectively.

Functions described in Eqs. (3) and (4) have been plotted in Fig. 4 together with their sum. Only two free parameters have been used in order to match the calculation and photoluminescence results, i.e., the proportionality coefficients between the excitation power and the generation rate (related to the optical injection efficiency which is smaller than 1 and

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unknown), and the one between $I_{X(XX)}$ and $p_{X(XX)}/\tau_{X(XX)}$. Both the coefficients shift the theoretical curves horizontally or vertically (in a double logarithmic scale) while not changing their slope or shape. The only semi-free adjustable parameter which influences the mathematical character of these dependencies is the ratio of the exciton to biexciton radiative lifetimes τ_X/τ_{XX}. In a very general and simplest quantum dot exciton energy level picture, two theoretical limits of this ratio can be considered, depending on the quantum confinement strength (after Ref. 8, for instance). In the strong confinement limit, the exciton to biexciton lifetime ratio is expected to be approximately 2 (explained by twice larger number of decay channels for biexciton) and should decrease significantly when going into the weak confinement regime.[8] There have been used $\tau_X/\tau_{XX}=2$ in Fig. 4 as the first approximation (shown with dashed lines), which seems to describe our experimental result quite well.

Similar exciton and biexciton PL intensity dependencies have been observed for several other quantum rods within this ensemble, and all of them show the same tendency. They are impossible to be explained with $\tau_X\tau_{XX}<2$ (confirming the rods are in the strong confinement regime rather), and actually a better agreement between the experiment and our model can be obtained for τ_X/τ_{XX} significantly larger than 2. One of the possible explanations to be considered was the contribution of higher order states occupation at high excitation/generation rate (neglected in our model). Nevertheless, when there was used an extension of the model into higher order states (as in Ref. 9), no improvement of the agreement to the experimental data has been obtained suggesting that the observed high excitation behavior is most probably not related to the contribution of the higher order levels.

Large τ_X/τ_{XX} ratios, which mean biexciton radiative lifetime significantly shorter than the exciton one, have been previously suggested in both theoretical predictions[8,10,11] and experimental reports[12,13] for different dots showing that they can occur under certain circumstances (geometries, contents, sizes, etc.). One of the fundamental reasons, proposed in a recent paper by Narvaez *et al.*,[14] is related to the existence of the exciton fine structure (i.e., a splitting into two bright and two dark states caused by electron-hole exchange interaction). The internal dynamics between these states has been shown to play a crucial role and the ratio of exciton and biexiton lifetimes (driven mostly by the exciton lifetime change) is controlled by the rate of spin-flip relaxation from bright to dark exciton states. Two limits can be distinguished: slow spin-flip case, which gives $\tau_X/\tau_{XX}=2$ and is typically observed in self-assembled InGaAs/GaAs QDs, and the other one for fast spin-flip, which gives $\tau_X/\tau_{XX}=4$ and has been observed in II–VI colloidal dots. Therefore, in Fig. 4, the theoretical dependencies for $\tau_X/\tau_{XX}=4$ are plotted (continuous lines), which give better agreement with the experiment. This might indicate that in case of the investigated InGaAs/GaAs quantum rods (high aspect ratio dots), in contrast to the observations for self-assembled InGaAs dots, rather fast spin-flip relaxation from bright to dark exciton states occurs. However, a more direct experimental confirmation, such as exciton and biexciton radiative lifetimes measurements, is needed.

The observed *X* and *XX* emission lines from several single quantum rods have allowed deriving a biexciton binding energy in the range from about 1 meV (as in the case presented in Fig. 3 where it is equal to 1.1 meV) to 2.2 meV. Such biexciton binding energy dispersion is related to the rod size, or even more probably to the In composition dispersion within the ensemble. The obtained binding energies are close to those previously observed and reported for typical self-assembled InAs or InGaAs quantum dots in GaAs host.[8,11,12,15] This can be understood taking into account that both the self-assembled dots and the quantum rods investigated here are in the strong confinement regime (even if at its large size limit). Indeed, in this range of sizes (volumes) a very weak dependence of the biexciton binding energy on the object size is expected[8,11] and can be fully dominated by other local ensemble properties.

In conclusion, there have been investigated photoluminescence spectra from single InGaAs epitaxial quantum rods with a high aspect ratio. The emission of an exciton and biexciton has been recognized and described by a rate equation model, suggesting that the radiative lifetime of the biexciton is more than twice shorter with respect to the exciton one, which confirms the quantum confinement to be in the regime of a strong confinement (but at its large size limit) and can be an indirect indication on fast bright to dark spin-flip relaxation rate in quantum rods. The detected exciton binding energies have been found to be similar to those for self-assembled dots, which is attributed to the weak dependence of the *XX* binding energy on the dot size in this range of confinement volumes.

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