One-dimensional features of In(Ga)As/GaAs dot chain structures with changeable interdot coupling

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New Journal of Physics 11 (2009) 043022 (9pp)
Received 5 December 2008
Published 15 April 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/4/043022

Abstract. In this paper, In(Ga)As/GaAs quantum dot chain structures with adjustable interdot coupling are studied by means of atomic force microscopy, excitation power density-, temperature-dependent- and time-resolved photoluminescence (PL) spectroscopy. According to the growth parameters, samples in this set model the transition from a 0D to a 1D system. It is shown that an interval of lattice temperatures exists where exciton dynamics is typical for the 1D system. In this temperature interval, the radiative decay time depends linearly on $\sqrt{T}$ in all the samples. The exciton dynamics is substantially affected by in-chain interdot distances that determine the interdot coupling and define the hopping character of the exciton thermal relaxation in In(Ga)As/GaAs dot chain structures.

The recent development of nanotechnology has allowed the assembling of various quantum dots (QDs) and nanocrystal structures possessing highly different physical properties for a wide range of applications [1]–[3]. With different spatial arrangement of the dots, it is possible to bring them into coupling and coherence in order to create an artificial medium having unique properties [4]. For example, due to the coupling of semiconductor QDs, ‘artificial molecules’ can be formed containing two or more QDs [5]–[7] with tunable interdot coupling. The strength of interdot coupling in the case of vertically stacked QDs is controlled by introducing spacer

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layers of different thicknesses between the layers of QDs. The natural limitation in such stacked QD structures is the formation of defects due to strain accumulation. In order to avoid this limitation, a lateral arrangement of QDs has been suggested. Thus, fabrication of planar structures containing various configurations of laterally coupled dots has been reported [8]–[10]. One example of this is that long chains (up to 5 µm in length) of uniform, dislocation-free In(Ga)As/GaAs QDs have been grown by means of molecular beam epitaxy (MBE) [11]. It has been demonstrated [11] that by changing the MBE growth procedure the QDs will form chains with predictable densities and sizes, which offers the possibility of engineering the interdot distances and hence the interdot coupling. In fact, changing the interdot distances within the chains facilitates the controlled transition from a zero-dimensional (0D) system to a 1D system (QDs to quantum wires (QWrs)). The study of the evolution of the physical properties of the In(Ga)As/GaAs dot chain structures under such a transition should provide us with a better understanding of the mechanism of interdot coupling and its role in the formation of the electronic states and carrier relaxation rates. Using atomic force microscopy (AFM), excitation power density-, temperature-dependent- and time-resolved photoluminescence (PL) spectroscopy, we present direct evidence of the availability of the 1D character of the carrier relaxation and its evolution during the transition from weak interdot coupling to the strong coupling regime in the In(Ga)As/GaAs dot chain structures.

The In(Ga)As QD chains were grown by MBE on semi-insulating GaAs (100) substrates. Following a 0.3 µm GaAs buffer layer grown at 580 °C, a layer of InxGa1-xAs QDs with a 60 ML GaAs cap was repeated 15 times at 540 °C to form a superlattice. Growth using uncracked As4 was performed using a constant beam equivalent pressure V/III ratio of 15:1. Several sets of InxGa1-xAs QD superlattices were grown with x equal to 0.3, 0.4 and 0.5 and QD layer thicknesses of 15.5, 8.5 and 5.7 MLs, respectively. These thicknesses were chosen to be 25% above the critical thickness for the 2D–3D transition for each composition as determined by in situ reflection high-energy electron diffraction. A final QD layer was grown and left uncapped for morphology analysis in each sample. The surface morphologies of the samples were analyzed by AFM to uncover distributions in the shape, size and lateral arrangement of the QDs. Figure 1 shows the AFM images of three representative samples. One can see long dot chains oriented along the [0-11] direction, with the spacing between chains significantly larger than the dot spacing within each chain. The in-chain interdot distance is varied realizing conventionally weak (figure 1(a), sample C1), intermediate (figure 1(b), sample C2) and strong (figure 1(c), C3) interdot coupling. The results of the AFM analysis are compiled in table 1. The
Table 1. QD parameters of In(Ga)As/GaAs dot chain structures derived from AFM analysis ($d_{[011]}$ and $d_{[0-11]}$ represent the average dot spacing in the orthogonal directions).

| Sample | $x$(In$_x$Ga$_{1-x}$As) | $d_{[011]}$ (nm) | $d_{[0-11]}$ (nm) | Height (nm) |
|--------|--------------------------|-----------------|-----------------|-------------|
| C1     | 0.5                      | 86              | 65              | 2.5         |
| C2     | 0.4                      | 83              | 44              | 5.5         |
| C3     | 0.3                      | 102             | QWR-like        | 3.0         |

QDs are slightly elongated along the chain direction of [0-11], and have average spacings and heights as given in table 1. It is clear from figure 1(a) that sample C3 represents a perfect QWR structure.

In order to reveal the effect of interdot coupling on carrier relaxation in the QD system, PL measurements were carried out in variable conditions. For continuous wavelength (cw) PL measurements, QDs were excited with the 532 nm line from a Nd:YAG laser. The laser spot diameter was ~30 µm and the optical excitation intensity was varied in the range of ~($10^{-5}$–1000) W cm$^{-2}$. The PL measurements were performed in a variable temperature 8–300 K closed-cycle helium cryostat. The PL signal from the sample was dispersed by a monochromator and detected by an LN-cooled InGaAs photodiode detector array. The transient PL was excited using 2 ps pulses at $\lambda_{exc} = 750$ nm from a mode-locked Ti:sapphire laser producing an optical pulse train at 76 MHz with excitation density varied between $10^9$ and $2 \times 10^{14}$ photons pulse$^{-1}$ cm$^{-2}$. A Hamamatsu synchroscan streak camera C5680 with an infrared enhanced S1 cathode was used for signal detection. The overall time resolution of the time-resolved PL system was ~15 ps.

The normalized PL spectra measured at $T = 10$ K and excitation intensity $I_{exc} = 0.4$ W cm$^{-2}$ are shown in the inset of figure 2(a). The PL spectra were fitted using a Gaussian profile with $E_{me1}^1 = 1.256$ eV, $\Gamma_1 = 53$ meV; $E_{me2}^2 = 1.247$ eV, $\Gamma_2 = 47$ meV; and $E_{me3}^3 = 1.265$ eV, $\Gamma_3 = 31$ meV for the energy of the PL band maximum, $E_{me}$, and full-width at half-maximum (FWHM), $\Gamma$, in samples C1, C2 and C3, respectively. The sample with the narrowest FWHM is the wire-like, C3 sample, which indicates that this sample has the narrowest QD size distribution. However, the strain fields in and around the self-organized QDs result in a piezoelectric contribution to the PL which is sensitively dependent on the QD geometry, average InGaAs composition and the In/Ga distribution profile [12]–[14]. The piezoelectric fields in turn affect the electronic properties of the In(Ga)As/GaAs QDs shifting the energy of the QD exciton transitions to the red. Thus, it has been shown in [15] that the piezoelectric fields blueshift the PL spectra of the elongated In(Ga)As islands as the excitation intensity is increased. This shift arises due to screening of the internal piezoelectric fields and thus flattening of the local bandstructure by photo generated carriers. Therefore, in order to understand how the piezoelectric field is contributing to the PL line shape, the PL measurements were carried out with varying excitation density. Figure 2 demonstrates the change of the PL FWHM and the shift of the PL maximum in three samples with different interdot coupling through a range of excitation intensities. The largest PL blueshift (~18 meV) and simultaneous narrowing (~7 meV) are observed in the sample with weak interdot coupling. Such a behavior of the PL emission is typical for the screening of internal fields by photo-excited carriers. This implies that the strength of the piezoelectric field is largest in the QD sample with weakly coupled QDs,
Figure 2. (a) Change of the FWHM and (b) shift of the PL band maximum, $(E_{me}(I_{exc}) - E_{me}(I_{min}))$ versus relative excitation intensity $I_{exc}/I_0$ for samples C1, C2 and C3. $I_0 = 1000 \text{ W cm}^{-2}$. Inset: low-temperature PL spectra for samples C1, C2 and C3 measured at $I_{exc} = 0.4 \text{ W cm}^{-2}$.

whereas in the wire-like QD sample the field is significantly smaller, leading to substantially less pronounced but still observable changes of the peak position and linewidth. Further increase of the excitation power density results in a continued blueshift of the peaks and an increase in their linewidths. This is attributed to the state filling effect that results in appearance of the excited QD states. In our further study, we use the excitation density corresponding to $I_{exc}/I_0 = 10^{-3}$ that falls onto the region of plateau both in figures 2(a) and (b). At this excitation intensity we believe that the piezoelectric fields are fully screened and the PL line shape is assigned mainly to the QD size distribution in all samples.

To further characterize this line shape and the carrier relaxation and radiative recombination times, the temperature dependence of the cw and transient PL was studied. It was found that as the temperature increases from 10 K up to $\sim 120$ K the PL band maximum, $E_{me}$, rapidly shifts to the red and the FWHM decreases in all samples. Such behavior is typical for QD ensembles exhibiting interdot carrier transfer [16]–[21] and results from the thermal transfer of carriers from smaller QDs to larger QDs as the temperature is increased. This
Figure 3. PL transients measured at two different temperatures for (a) sample C1, (b) sample C2 and (c) sample C3. Solid lines show parts of mono-exponential dependences.

leads to an FWHM reduction and redshift. The increase in the QD FWHM detected at higher temperatures and the concomitant reduction in the rate of decrease of the $E_{me}$ energy is interpreted as the consequence of the broadening of the carrier distribution function with an increase in temperature. The mechanisms of low-temperature carrier transfer are widely discussed [16], [22]–[24] and include electron tunneling based on coupling between neighboring QDs following a semiclassical Wentzel–Kramers–Brillouin approximation [20]; transitions through continuum states that are assumed to be located in the vicinity of QDs [22, 23]; and resonant energy transfer via an optical near-field interaction [24].

Figure 3 demonstrates the behavior of the PL transients measured at the PL band maximum at two different temperatures for our samples. Solid lines represent a mono-exponential decay giving the PL decay time, $\tau_d$, according to $I_{me}(t) = I_{me}(0) \exp(-t/\tau)$. Time $t = 0$ ps corresponds to the choice of the time origin as the beginning of the decaying part of the
Figure 4. (a) Dependence on temperature of the PL decay time, measured at the maximum of the PL band and (b) integrated PL intensity in samples C1, C2 and C3.

$I_{me}(t)$ dependence. One can see an increase of the $\tau_d$ value in all samples with an increase in temperature. At temperatures higher than those shown in figure 3 the $\tau_d$ values rapidly decrease. Figure 4 summarizes the decay times, $\tau_d$, (figure 4(a)) together with the temperature dependence of the integrated PL intensity $I_{int}(T)$ (figure 4(b)) in all samples. Here, we see dramatic correlations between $\tau_d(T)$ and $I_{int}(T)$. As the temperature increases a growth in $\tau_d(T)$ is observed, while $I_{int}(T) \approx$ const for all samples. Further increase in temperature results in a rapid decrease in both $\tau_d(T)$ and $I_{int}(T)$. This later correlation is very important for understanding the mechanisms that govern $\tau_d(T)$. From an Arrhenius plot of $I_{int}(T)$ we estimate the QD exciton binding energy to be $\sim 12$ meV in all our samples.

The stability of the integral PL intensity with temperature for QD ensembles indicates the absence of non-radiative processes at low temperatures. This means that carriers or excitations are transferred in the QD ensemble and can recombine radiatively only in the QD system. At higher temperatures the carriers can escape from the QD ensemble and disappear in the wetting layer, traps and/or the GaAs barrier, thus reducing the integrated PL. In order to understand the character of the carrier transfer in the dot chain samples at low temperatures, we analyze $\tau_d(T)$ just in the range of temperatures where $I_{int}(T)$ remains constant. Following [25]–[27], the $\tau_d(T)$
functions were re-plotted versus $\sqrt{T}$, as shown in figure 5. In this case, the relevant parts of the curves can be obviously identified by the linear dependence on the $\sqrt{T}$ with the coefficients shown in figure 5. In order to interpret this, we used the results of the analysis given in [26] for exciton thermalization in a 1D (QWrs) system.

At low temperatures this thermalization is characterized by the parameter $\zeta(T)$, which represents the fraction of excitons with $k < k_0$, where $k_0$ is the wave vector of light in the QD sample with the same energy as the exciton. In the case of the Maxwell–Boltzmann distribution for excitons, $\zeta(T)$ can be presented as [26]

$$\zeta(T) = \frac{1}{\sqrt{\pi k_B T}} \int_{0}^{\Delta} D(E) \exp(-E/k_B T) \, dE \int_{0}^{\infty} D(E) \exp(-E/k_B T) \, dE, \quad (1)$$

with $D(E)$ being the density of states (DOS) for excitons and $\Delta = \hbar^2 k_0^2 / 2M$, where $M$ is the exciton mass being the maximum kinetic energy of excitons that can decay radiatively. For the case of the 1D system, 1D DOS $\propto 1/\sqrt{E}$. Thus, assuming that $T \gg \Delta / k_B$, one obtains

$$\zeta_{1D}(T) = \sqrt{\frac{4\Delta}{\pi k_B T}}. \quad (2)$$

The average radiative lifetime will then be

$$\tau(T) \cong \tau_0 \zeta(T)^{-1} = \tau_0 \sqrt{\frac{\pi k_B}{4\Delta}} \sqrt{T}, \quad (3)$$

with $\tau_0$ being the intrinsic radiative lifetime of the exciton at $k \sim 0$. Therefore, the radiative lifetime $\tau(T)$ is proportional to $\sqrt{T}$ for a 1D exciton system.

In terms of equation (3), the low-temperature behavior of $\tau_d(T)$ in sample C3 follows the behavior of the exciton lifetime $\tau$ in a 1D system with the coefficient of 96 ps K$^{-1/2}$ being close
to the 140 ps $K^{-1/2}$ [25] and 92 ps $K^{-1/2}$ [26] found in GaAs QWrs and the 102 ps $K^{-1/2}$ found in [27]. As can be seen from figure 5, the $\tau_d(T)$ dependence in sample C3 is practically flat up to $\sim 30$ K. Thus, in the temperature range 10–30 K the excitons are localized inside the QDs, and above 30 K they move freely through the QD chain as in the 1D energy band. Also, in this region, there is no loss of the integrated PL intensity (figure 4(b)), so, ultimately, the excitons must find their way to a dot to recombine. In the case of sample C2, the $\sqrt{T}$ coefficient equals 326 ps $K^{-1/2}$. This larger value indicates that interdot distances are larger in sample C2 than in sample C3. Due to the larger interdot distance, the overlap of electron wavefunctions of adjacent QDs is smaller; thus the carrier transfer is likened to interdot hopping. The temperature interval where the QD excitons stay localized is wider in this sample, from 10 K up to 40 K. Above this interval, the carrier transfer has a 1D character. This tendency is also traced for sample C1, with the weakest for the sample set interdot coupling. Here, the linear coefficient takes the value 472 ps $K^{-1/2}$, which indicates that interdot distances are the largest. It is important that even in sample C1 the 1D character of the exciton thermalization is preserved. The temperature interval of QD localization spreads from 10 K up to 60 K.

Finally, the set of In(Ga)As/GaAs dot chain structures with changeable interdot coupling is studied by means of AFM, excitation power density-, temperature-dependent and time-resolved PL spectroscopy. The set of samples allows realization of weak (0D), intermediate and strong (1D) interdot couplings. It is found that the exciton PL spectra depend on the excitation power demonstrating a blueshift and FWHM narrowing that is typical for piezoelectric field screening by photo-excited carriers. It is demonstrated that there exists a low-temperature interval depending on the strength of interdot coupling where the exciton dynamics is typical for 0D systems. In this temperature interval, radiative decay time does not depend on the temperature. It is shown that an interval of temperatures exists where the exciton dynamics is typical for the 1D system. In this temperature interval, the radiative decay time depends linearly on $\sqrt{T}$ in all the samples. The exciton dynamics is substantially affected by in-chain interdot distances that determine the interdot coupling and define the hopping character of the exciton thermal relaxation in In(Ga)As/GaAs dot chain structures. These results give deeper insight into the physics of coupled QDs important for various optoelectronic applications.

Acknowledgment

We acknowledge the US National Science Foundation financial support through grant no. DMR-0520550.

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New Journal of Physics 11 (2009) 043022 (http://www.njp.org/)
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