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2009 J. Phys.: Condens. Matter 21 025302

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IOP Publishing — Journal of Physics: Condensed Matter
J. Phys.: Condens. Matter **21** (2009) 025302 (4pp) doi:10.1088/0953-8984/21/2/025302

Carrier transport investigation in short-wavelength InAs/AlGaAs quantum dots

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Received 2 September 2008, in final form 28 October 2008
Published 9 December 2008
Online at stacks.iop.org/JPhysCM/21/025302

Abstract
Spatially resolved photoluminescence has been used to investigate the details of the carrier capture and recombination dynamics in InAs/AlGaAs self-assembled quantum dots. The spatial PL distribution displays a Gaussian-like profile, whose width depends upon the temperature and detection energy being analyzed. The results give evidence of carrier thermalization between dots with different sizes. The effects of carrier transport in the quantum dot (QD) structure and carrier capture cannot be separated. The results can be modeled by assuming a carrier hopping process.

Self-assembled semiconductor quantum dots (QDs) are very important widely for their application in novel optoelectronic devices [1–5]. In this context, the red-emitting InAs/$Al_xGa_{1-x}As$ quantum dot system is particularly interesting because of its applications requiring shorter wavelengths [2].

The overall migration of the excitation is a complex process in a nonuniformly excited medium. It is a cumulative result of ambipolar diffusion of electron–hole pairs, exciton diffusion, radiative transport, photon recycling, and tunneling, modified everywhere by the capture and possible thermal re-emission from the QDs [4–10]. The net result is a spreading of the excitation that can be experimentally monitored by observation of the specific emission generated by the electron–hole recombination in the QD ground and excited states.

We have used a spatially resolved photoluminescence (PL) technique to investigate the temperature dependence of the recombination kinetics of InAs/$Al_{0.6}Ga_{0.4}As$ dots. Carrier diffusion varies with the temperature and also with the detection energy (E_{det}). The results are modeled by assuming a carrier hopping process between different dots [5]. Previously, time-resolved experiments have shown that the carrier capture into the dots is fast and efficient despite the indirect nature of the barriers, which also explains the strong red emission from the samples [5].

Thermally activated carrier hopping is responsible for the decrease of the capture rates in the subset of small QDs (high energy) and increase in the subset of large dots (small energy). The analysis has also been confirmed by CW measurements in terms of the narrowing of the PL spectrum with the temperature and the luminescence peak redshift larger than the thermal shrinkage of the InAs bandgap [7, 8].

The InAs–$Al_{0.6}Ga_{0.4}As$ SAQD samples were grown by the Stranski–Krastanow technique using molecular beam epitaxy (MBE) on a [100]-oriented n^+-GaAs substrate. Following the GaAs substrate the sample structure has the following sequence: 200 nm GaAs buffer layer, 200 nm $Al_{0.6}Ga_{0.4}As$ barrier, 2.4 ML InAs QDs grown at 500 °C with a growth rate of 0.1 ML s^{-1}, and 100 nm $Al_{0.6}Ga_{0.4}As$ barrier. The structure was terminated with a 5 nm GaAs cap-layer.

A schematic representation of the micro-PL experiment is shown in figure 1. A CW argon-ion laser beam working at average power of 10 mW is focused onto a spot of about 3 μm on the sample surface using a microscope objective with long working distance. Emission from the sample is collected backward by the microscope objective. A beam-splitting prism is used to reflect the light back from the sample towards the scanning monochromator. The spatially resolved PL measurements were performed from 10 to 160 K at two excitation lines corresponding to 514 nm (E_{ex} = 2.41 eV) and 457 nm (E_{ex} = 2.71 eV) of an Ar^+-ion laser. The magnified image was scanned using a pinhole attached

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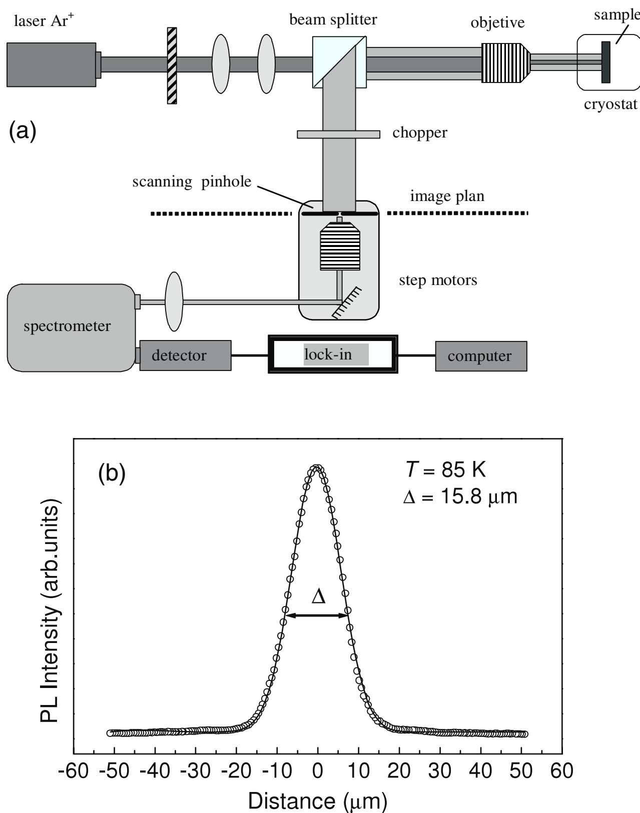

Figure 1. (a) Schematic representation of the micro-PL surface scan technique. (b) Typical PL image obtained by scanning the PL on the sample surface.

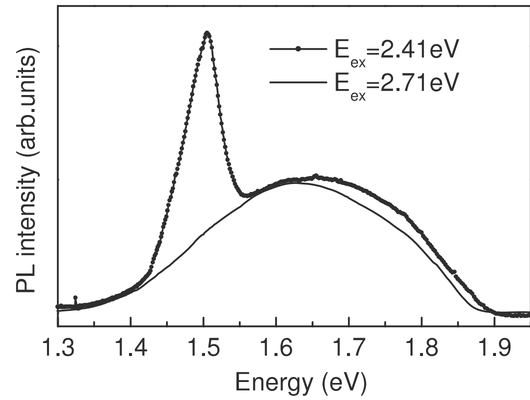

Figure 2. PL spectra measured at two excitation energies at 10 K.

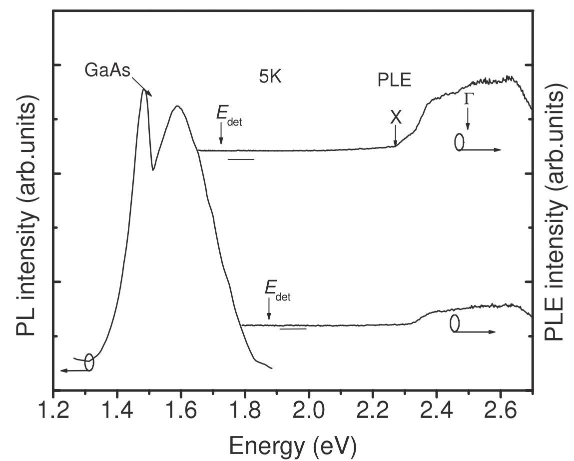

Figure 3. PLE spectra measured at two detecting energies on the QD PL emission.

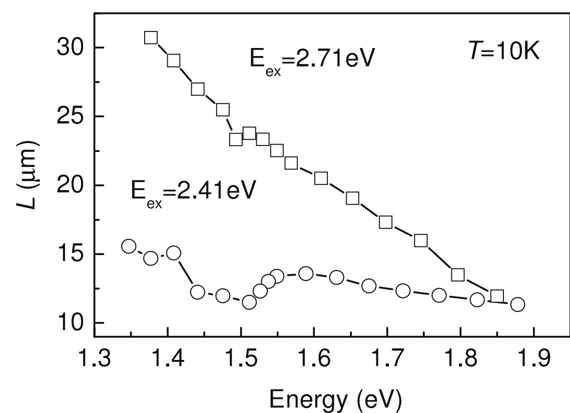

Figure 4. Carrier diffusion parameter L as a function of the detection energy measured at two excitation energies.

to micro-step motors. The emitted light was resolved by a 0.5 m monochromator and detected by a nitrogen-cooled Ge detector using lock-in detection. A typical spatially resolved PL is demonstrated in figure 1(b). The result we observe is a spreading of the radiative recombination that can be experimentally monitored by observation of the specific emission generated by the electron–hole pair recombination in the ground and excited states of the QDs.

Figure 2 shows the PL spectra of the InAs/$Al_xGa_{1-x}As$ QDs. The QD emission is centered at about 1.65 eV. Emission from the GaAs layers can be seen on the left-hand side of the PL when the sample is excited with the green line at 514 nm ($E_{ex} = 2.41$ eV). The non-resonant excitation energy generates carriers predominantly in the Γ-valleys of the AlGaAs barriers as observed in the PLE spectra in figure 3. The PL spectra demonstrates that the GaAs emission is stronger compared to the AlGaAs when the structure is excited with the green line. When exciting with the blue line ($E_{ex} = 2.71$ eV) it is hard to distinguish the emission from the GaAs layer, since the emission in the AlGaAs layer is enhanced by a factor of 50% approximately.

The spatial PL distribution displays a Gaussian-like profile, such as that in figure 1(b), whose width at half maximum Δ depends basically on the carrier diffusion length L, that is strongly correlated to the temperature and detection energy being analyzed. The carrier diffusion parameter L was then measured as a function of the detection energy for two excitation energies as in figure 4. This parameter is quite useful and has been correlated with conventional diffusion lengths in 2D and 3D systems, despite being a simplification of the transport problem [6].

Figure 4 demonstrates that, depending on the excitation energy used, we are able to control the carrier diffusion lengths in the QD structure. Then, exciting the sample with the blue

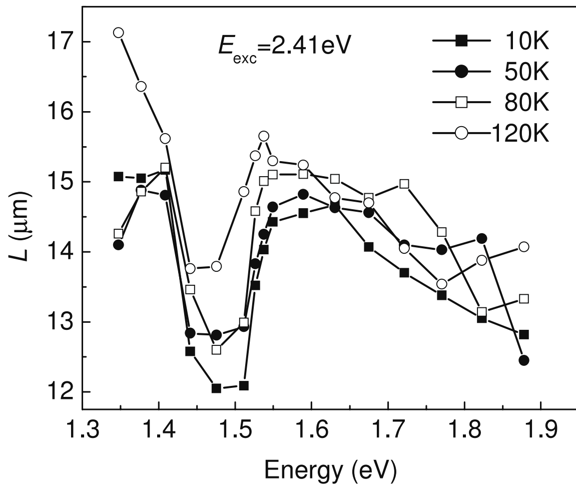

Figure 5. Carrier diffusion parameter as a function of the detection energy obtained at different temperatures.

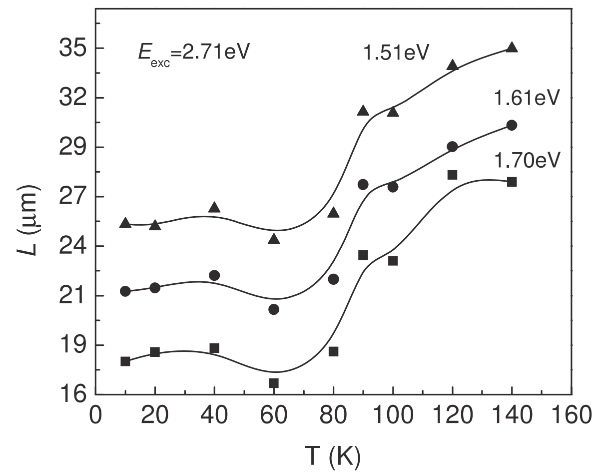

Figure 6. Carrier diffusion parameter as a function of the temperature measured for three detection energies.

line we have longer diffusion lengths, since the density of carriers being captured by the dots through the AlGaAs barriers is higher. We also measure an effective diffusion length that can be different for different QD sizes. Generally, the carrier density in different energy levels of QDs will change with position.

The PL under non-resonant excitation is determined by carrier capture into the dots and carrier relaxation inside the dots. The majority of the electron–hole pairs are created in the GaAs and AlGaAs barriers from which they are fast captured by the QDs on a timescale of approximately picoseconds [6]. Once the carriers are captured by the QDs, it is generally accepted that they relax sequentially via the excited states to the ground states. We describe the overall process in terms of a carrier migration length that we assume is primarily determined by the ambipolar diffusion equation, but in many cases it is modified by the local capture and recombination of carriers in the wetting layer and QDs.

There is significant migration of electron–hole pairs outside the illuminated spot. Because of photogeneration of carriers in the surrounding barrier material, lateral migration occurs primarily due to carrier diffusion in AlGaAs accompanied by the capture of carriers into the barriers and ultimately into QDs. This leads to the lateral spreading of the emission spot. The emission intensity in these cases is controlled by the local capture rate of carriers into QDs with a subsequent distribution over available local states.

According to figure 4, there is a drastic decrease of the diffusion length around the energy of 1.47 eV. Another behavior is seen exciting with the blue line ($E_{ex} = 2.71$ eV). To understand such behavior, we followed its temperature dependence. We had to follow its temperature dependence in order to be sure that this behavior is not linked to defects. The thermal effect could activate or deactivate the defects.

The most important steps for diffusion started from the carrier capture into the dots. In figure 5, the temperature dependence of L shows the increase with temperature on the low-energy side of the PL, although the diffusion lengths do not have the same dependence on the high-energy side. It is also expected that the diffusion is larger at higher temperatures followed by thermally activated energy processes. Therefore, the carrier diffusion inside the QD structure is strongly suppressed by the capture into the dots at low temperatures. Any hypothesis that the carrier diffusion quenches at approximately 1.47 eV must also take into account the increase of the radiative recombination in the GaAs layer. Any influence of tunneling between dots on the lateral migration is not seen and can be disregarded.

In time-resolved measurements, the PL rise time under non-resonant excitation was determined by carrier capture into the dots and carrier relaxation inside the dots [5]. It was demonstrated that the majority of the electron–hole pairs are created in the Γ-valley of the $Al_{0.6}Ga_{0.4}As$ barriers, and then they scatter rapidly to the X-valley, from which they are captured by the QDs on a timescale of approximately 50 ps at high power and low temperatures. So, the process of carrier diffusion should take into account the energy transfer of excitation from the laser spot to the surrounding dots. Excitation transfers from high-energy levels are more absorbed; in contrast, excitation transfers from low-energy levels are less absorbed and for this reason the energy spreading may be larger. When the green line ($E_{ex} = 2.41$ eV) is used for excitation, the luminescence intensity is stronger around 1.5 eV, which corresponds to the GaAs emission. The emission energy is strongly absorbed by the surrounding dots, especially the dots in resonance and transferring energy. The dots are consequentially full and the energy transfer into these dots is bleached and the spreading is reduced. This behavior is controlled by the ambipolar diffusion of carriers in the GaAs and by recombination in the QDs. The saturation of the QD emission can also be demonstrated.

Figure 6 depicts the temperature dependence of the parameter L when the QDs are excited with the blue line. The increase of L is more pronounced above 80 K. We have chosen three detection energies to show that the increase occurs predominantly at all energies; the lower the energy

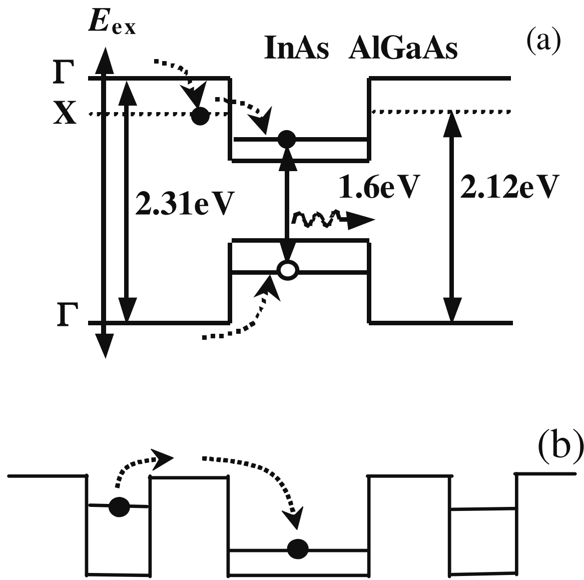

Figure 7. (a) Schematic representation of the band structure of InAs–AlGaAs QDs with indirect barriers. (b) Representation of the carrier hopping process between adjacent QDs that is considered in our model.

the higher the diffusion length. Most descriptions of the carrier population kinetics in self-assembled QDs assume capture of electron–hole pairs into the dots from an external reservoir (barrier material or wetting layer) with a steady-state occupation governed by the relative timescales for carrier capture (electrons and holes in equal numbers), relaxation, and radiative recombination [7]. To model our system, at low temperatures, carriers recombine from the full range of the dot states, and as the temperature increases they can diffuse more easily in the growth plane and occupy the lowest-dot-energy states. However, the overall migration of the excitation may be a complex process resulting mainly from ambipolar diffusion of electron–hole pairs (or excitons) and radiative transport, such as photon recycling [6, 9].

The effects of carrier transport in the QD structure and carrier capture into the dots cannot be separated. The measured rise times demonstrate that the rate of carrier capture into the dots increases with the temperature (high-energy side of the PL emission—in the temperature range of 5–293 K) [9]. For this reason, the carrier diffusion length in the QD structure should decrease with the temperature. This behavior is better observed by measuring the spatial PL distribution. For this reason a comprehensive understanding of the carrier dynamics can be obtained when the diffusion properties is compared with the capture times.

The temperature dependence of L shown in figure 6 suggests a process of thermal carrier distribution between dots. With increasing temperature the carriers can overcome the shallow energy minima by thermal activation and hop into deeper states. This process is illustrated in figure 7. The carriers are assumed to be frozen into the inhomogeneous distribution of QD states at low temperatures. With increasing temperature, carriers may be thermally activated outside the dot into the wetting layer and AlGaAs barrier and then relax into a different dot. The carrier hopping between dots then favors a drift of carriers toward the dots with the largest confining potential, and hence a lower-energy emission. Therefore, we expect a decrease of the capture times on the lower-energy emissions with the increase of the temperature due to the diffusion of carriers in the barriers.

In summary, we have carried out the study of carrier diffusion in InAs–$Al_{0.6}Ga_{0.4}As$ self-assembled QDs as a function of the temperature and detection energies using a confocal microscopy technique. The spatial scanning of the microluminescence in the focal plane allows us to obtain the distribution of the optical signal as a function of the radial displacement. Our results suggest that the capture processes are linked to the effects of carrier diffusion in these QDs.

Acknowledgments

We acknowledge financial support from Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq) and Fundação de Amparo à Pesquisa do Estado de Minas Gerais (FAPEMIG). We also acknowledge Professor M Skolnick for the sample used in this work.

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