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Interdot excitation transfer and spectral diffusion: consequences of wetting layer potential fluctuations in self-assembled quantum dots

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Abstract

A thin quantum well called the wetting layer is created whenever self-assembled quantum dots are fabricated. The wetting layer is highly non-uniform, thereby producing potential fluctuations that influence the physics of the quantum dots. Here, we report several effects that are caused by such fluctuations. In particular, light-induced spectral diffusion on a very long time scale and interdot excitation transfer are observed in a sample of InAlAs quantum dots. We show that the existence of both these phenomena is due to the wetting layer fluctuations.

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1. Introduction

Self-assembled quantum dots (SADs) are manufactured using conventional epitaxial techniques, by growing a semiconductor on a barrier material to which it is strongly lattice mismatched [1,2]. When the thickness of the epitaxial layer reaches a certain critical level, the strain is partially relieved by spontaneous formation of islands in the layer. If the growth is interrupted soon after this transition has occurred, the islands remain dislocation free and relatively uniform in size. The sample is then capped with barrier material, transforming the islands into quantum dots (QDs) that can confine carriers in all three spatial dimensions.

After the dots have formed, some of the QD material remains distributed in a two-dimensional wetting layer (WL) that connects all the SADs. The WL is highly non-uniform [3], which inevitably leads to potential fluctuations in the barrier separating the dots. These fluctuations can be quite large since the WL is typically quite thin (<20 Å), and occur in the immediate vicinity of the QDs. In spite of this, most studies to date neglect such effects, treating SADs as uncoupled, near ideal zero-dimensional (0D) systems. Here we describe several recent observations in a SAD sample that can only be explained by departing from the ideal model to take into account WL potential fluctuations.

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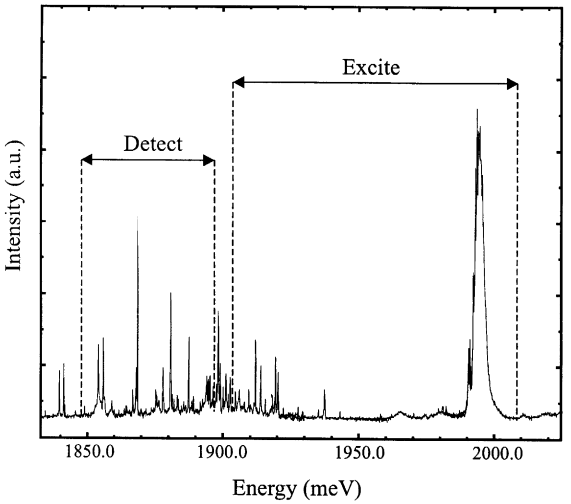

Fig. 1. A typical PL spectrum of the sample taken in the near-field. The sharp lines between 1840 and 1940 meV are due to individual quantum dots, and the feature at 1995 meV is the barrier exciton. Detection and excitation wavelengths for PLE are schematically indicated.

2. Experimental

The sample consists of an $In_{0.55}Al_{0.45}As$ quantum dot layer embedded in an $Al_{0.35}Ga_{0.65}As$ barrier. This sample is well characterized [4–7], with a dot density of $\sim$200 dots/mm^2, a lateral dot size of $\sim$18 nm, and a ground state–excited state splitting of 35–40 meV, which means that the ground state and two excited states are confined below the $\sim$140 meV lateral energy barrier. In order to resolve the spectral lines from individual SADs, a near-field scanning optical microscope (NSOM) capable of operating at low temperatures was used to limit data acquisition to a small area containing only a few quantum dots. A typical photoluminescence (PL) spectrum taken in the optical near field is shown in Fig. 1. The sharp lines between 1840 and 1940 meV are emission from the ground states of individual quantum dots, and the feature at 1995 meV is due to the AlGaAs barrier exciton.

2.1. Nature of wetting layer

The WL in the sample under investigation is so strongly disordered (due to interface and/or alloy fluctuation) that it contains no continuum of delocalized 2D states. This can be deduced from PLE spectra taken at energies below the barrier exciton, as shown if Fig. 2a. If the WL were a 2D quantum well, a resonance associated with the WL exciton would be visible in the spectrum. Instead, only a slowly decaying absorption tail extending well into the QD emission region can be seen. In addition, if one examines PL spectra at energies immediately below the barrier exciton, one can discern a number of very weak, easily saturable sharp lines, as shown in Fig. 2b. This, too, is clear evidence of the local nature of WL states in this sample.

2.2. Spectral diffusion

At low optical power, the PL spectrum is stable over time. However, if the tip output power is increased above approximately 2 μW, the energy of the emission lines begin to slowly shift as a function of time, as shown in Fig. 3. This phenomenon is known as spectral diffusion (SD). At sufficiently high powers, we observe SD at a very long time scale (minutes to hours) in virtually all QD lines.

A more extensive analysis of this experiment is published elsewhere [8], and here we only relate the main points of that discussion: Because of the very long time scale of the SD, trapping of charge near the dots is the likely cause. Since the sample is not in mechanical contact with the near-field tip, and other

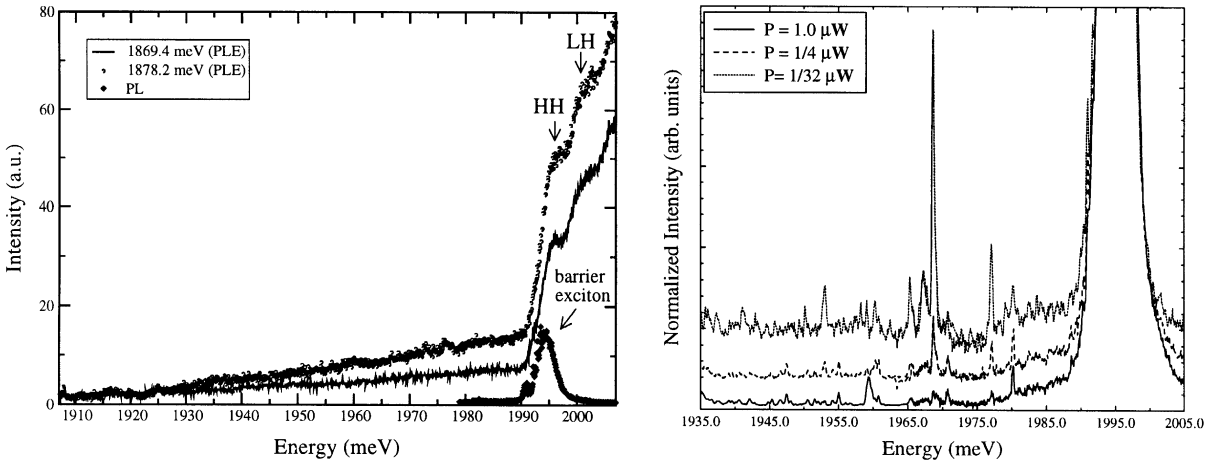

Fig. 2. (a) PLE spectra of two quantum dot emission lines superimposed with the PL of the barrier layer exciton. There are no vertical offsets of the graphs. No WL bandedge is visible down to 1907 meV. (b) PL with $\lambda_{exc} = 514.5$ nm. The spectra have been linearly scaled inversely to power and offset slightly to enable comparison. The heights of the barrier exciton peaks at 1995 meV are about eight times the full vertical scale of the figure. The sharp lines below the barrier are easily saturable, demonstrating the local nature of the WL states.

perturbations are unlikely to affect the structure of a stable semiconductor crystal, structural changes can be excluded. The fact that the SD is light induced leads to the same conclusion: light can generate charges in a semiconductor through several mechanisms, such as photoionization of excitons or auger recombination of biexcitons. The WL provides a sufficient number of local traps to serve as long-lived charged states to explain the observations. Since we are exciting at energies well below the AlGaAs barrier, these traps must lie in or about the WL or its interface with the barrier, where strong potential fluctuations are known to exist. Thus one effect of a non-uniform WL is light-induced spectral diffusion with long time scales.

2.3. Interdot excitation transfer

Photoluminescence excitation (PLE) spectroscopy was performed on the sample in the same experimental configuration as described in the previous subsection. The laser was tuned through wavelengths below the barrier exciton and WL energy to map out the excited states of individual dots. Resonances consistent with the second excited states of the dots were indeed seen in the data, as shown in Fig. 4. However, as is also clear from Fig. 4, resonances in sets of two or more SAD emission lines often occur at exactly the same excitation energy. The splitting between emission lines of such sets varies from between 0.5 and 25 meV, a range of energies inconsistent with the major state splitting ($\sim$35 meV) as well as any fine-structure splitting ($<$a few meV). The intensity of the lines is also linear in incident power both on and off resonance, excluding non-linear effects such as multiple excitons. Hence, each line in a set must originate from a separate quantum dot [9].

We then conclude that the cause of the phenomenon is transfer of excitation into a few nearby dots when a particular QD is directly excited, causing more than one dot to light up simultaneously. The set of dots exhibiting resonance is also much smaller (a few) than the number excited through ordinary diffusion from WL or barrier states (dozens). Thus interdot transfer occurs on a short length scale and at particular excitation energies, while regular diffusion occurs over a much larger length scale.

There are several mechanisms that could cause such an interaction. However, since the typical dot-to-dot separation is larger than 10 nm, only Förster [10] (dipole–dipole) interaction is sufficiently long range to transfer excitation directly over such a distance. The Förster interaction requires overlap between the donor emission spectrum and the acceptor absorption spectrum. Since all bound states in the SADs are quite

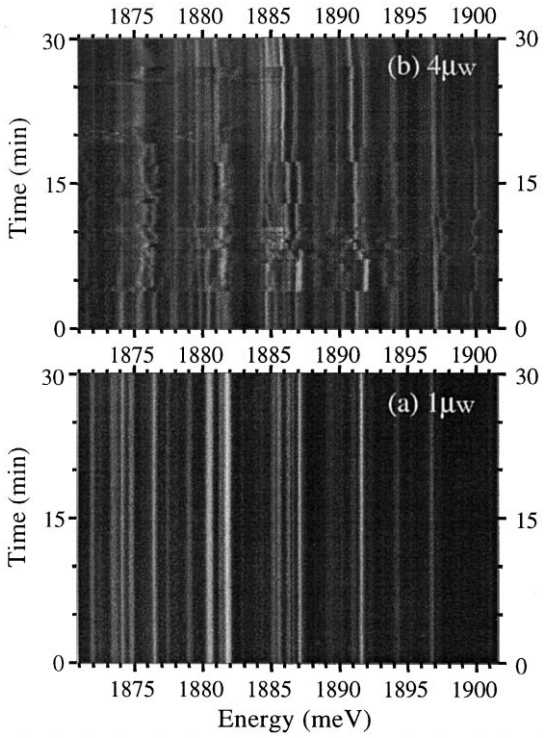

Fig. 3. Near-field PL spectra as a function of time and energy of a number of individual SAD emission lines. The gray scale indicates signal intensity. Total tip output power was 1 μW in (a) and 4 μW in (b). Spectral diffusion is not visible in (a), but clearly visible in (b).

narrow (<0.4 meV) and emission typically is seen only from the ground state, this condition is in general not satisfied. This implies that the lateral interdot excitation transfer is indirect in nature, and we again invoke the WL potential fluctuation to produce the necessary intermediate states. The precise mechanism for the transfer remains unknown, although multi-step exciton tunneling and multiple Förster-type energy transfers are two good candidates.

3. Conclusion

Spectral diffusion and interdot coupling show that the potential fluctuations of the WL have a strong influence on the physics of the quantum dots. Even though the severity of the fluctuations may be smaller

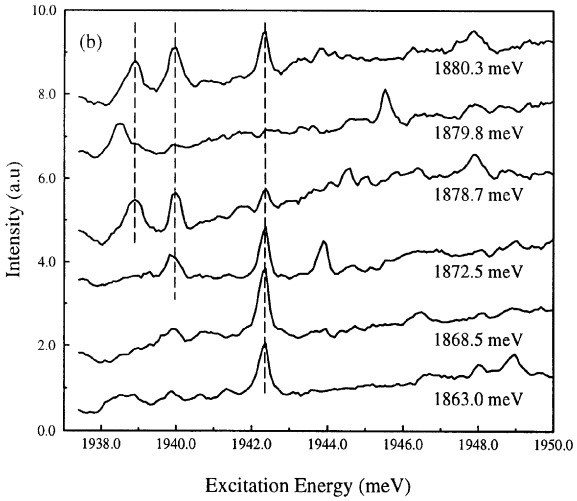

Fig. 4. Simultaneously acquired PLE spectra of different SADs. The label of each trace is the detection energy. The dashed lines indicate resonances occurring simultaneously in more than one dot. Traces are offset for clarity.

in other SAD samples, we believe that similar phenomena are occurring, albeit on different time- and length scales.

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