# LATERAL COUPLING OF SELF-ASSEMBLED QUANTUM DOTS STUDIED BY NEAR-FIELD SPECTROSCOPY.

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## ABSTRACT

Lateral coupling between separate quantum dots has been observed in a system of $In_{0.55}Al_{0.45}As$ self-assembled quantum dots. The experiment was performed by taking photoluminescence excitation (PLE) spectra in the optical near-field at 4.2 K. The high spatial resolution afforded by the near-field technique allows us to resolve individual dots in a density regime where interactions between neighboring dots become apparent. In the PLE spectra, narrow resonances are observed in the emission lines of individual dots. A large fraction of these resonances occur simultaneously in several emission lines, originating from different quantum dots. This is evidence of interdot scattering of carriers, which additional data show to be mediated by localized states at energies below the wetting layer exciton energy. A very rich phonon spectrum generated by the complicated interfaces between barrier and dot material is also evident in the data.

## INTRODUCTION

The study of self-assembled quantum dots (SADs) [1-5] is currently an extremely active field. This is due mainly to the high material quality and the relative ease of fabrication, involving no processing beyond the growth itself. At the same time, the study of this system presents a challenge, as the placement of individual dots is difficult to control, and the dot density can be quite high. Thus traditional experimental techniques often only allow simultaneous observation of large ensembles of quantum dots, where inhomogeneous broadening washes out many interesting features. Near-field scanning optical microscopy (NSOM) offers an improvement of this situation, as it enables optical microscopy and spectroscopy with spatial resolution beyond the Bragg limit [6]. This paper will discuss lateral coupling of quantum dots, caused by interdot scattering of carriers. This phenomenon requires a relatively high dot density in order to occur, and therefore cannot be directly observed with conventional optical techniques, although its existence can sometimes be indirectly inferred [7,8].

## EXPERIMENTAL

The sample used in this study consists of an $In_{0.55}Al_{0.45}As$ MBE grown quantum well embedded between $Al_{0.35}Ga_{0.65}As$ barriers. Due to the lattice mismatch between the two materials, the strain is relieved during growth by the formation of islands in the quantum well, resulting in a disordered array of quantum dots laterally connected by the quantum well, which is also known as the wetting layer (WL). This particular sample used here has previously been studied extensively [9-13], and the dot density is known to

be $2 \cdot 10^{10}$ $cm^{-2}$, the average dot radius is approximately 9 nm, and the ground state and the first excited state are separated by about 40 meV.

A low temperature NSOM was used to perform localized spectroscopy on the sample, the high spatial resolution achievable by this instrument allowing observation of an ensemble of quantum dots sufficiently small that the spectra of individual dots can be readily discerned. The measurements were performed in the so called as illumination mode, where the exciting radiation is delivered through the aperture of a near-field tip, and the photoluminescence (PL) signal is collected using conventional far-field optics. While this mode of operation is the simplest to execute and yields the largest signal, it has the disadvantage that the spatial resolution is degraded by the diffusion of carriers away from the point of excitation. In our experiment, however, the excitation energy is mostly kept below the energy of the wetting layer free exciton, where only localized states are available, reducing carrier diffusion by more than a factor of 2.

PLE measurements were carried out by detecting the emission from several quantum dots in parallel while the excitation source was tuned from wavelengths just above the wetting layer exciton energy to the dot emission wavelengths (Schematically illustrated in Fig. 1).

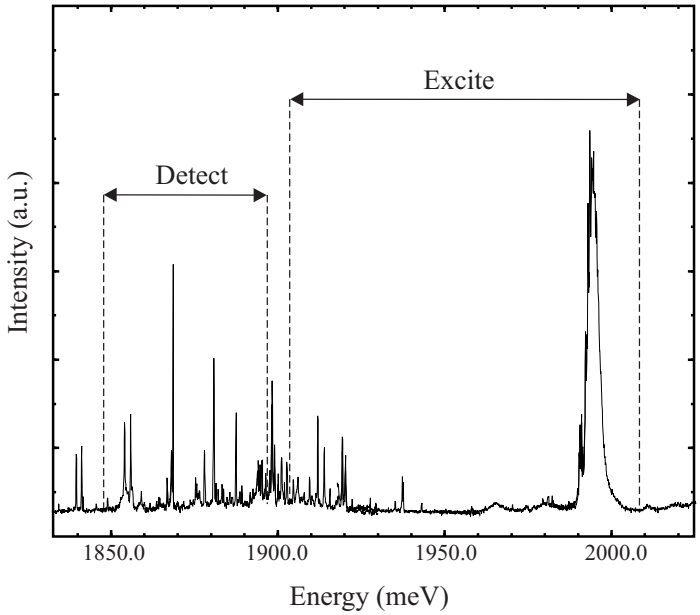


**Figure 1.** A typical PL spectrum of the sample taken in the near-field. The sharp lines between 1840 meV and 1940 meV are due to individual quantum dots, and the feature at 1995 meV is the WL exciton. Detection and excitation wavelengths for PLE are schematically indicated.

## RESULTS AND DISCUSSION

Fig. 2a plots the of emission in gray scale as a function of both excitation and detection energies. Each of the vertical lines corresponds to the emission from a single quantum dot. It is important to note that since the excitation energies are below the wetting layer exciton, we would *a priori* expect to see a PL signal only when the laser is resonant with an excited state in one of the local quantum dots. This is clearly not the case, as the PLE shows a strong non-resonant component in nearly all the emission lines, manifest in the figure as vertical lines. This is analogous to the situation in glasses and heavily doped semiconductors, where a tail in the density of states below the regular band

gap gives rise to similar behavior. In our case, the semiconductor is undoped, and we attribute the non-resonant PLE to localized states present below the WL band edge. This assignment is corroborated by PL data exemplified in Fig. 2b, where a number of sharp, easily saturable lines can be seen at energies just below the WL exciton. Moreover, it is known[14] that the WL in SADs is riddled with fairly large magnitude interface fluctuations, and likely to contain alloy fluctuations typical of such ternary alloys. Both these effects could generate the potential fluctuations required to explain the observed effects.

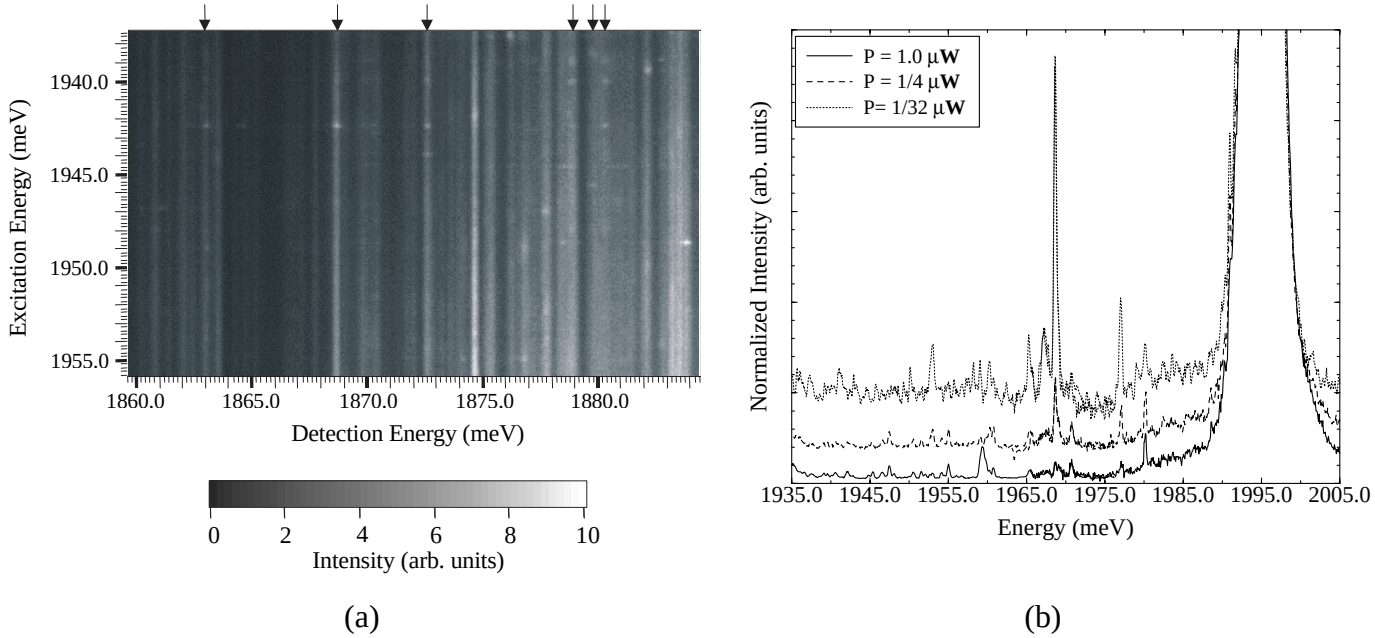


**Figure 2.** (a) Typical PLE scan plotting of intensity vs. excitation and detection energies. Numerous resonances are visible. The arrows mark the positions of the PLE spectra plotted in Fig. 3. (b) PL spectra from the energy region immediately below the WL, showing numerous localized sharp features. The intensity scale has been normalized to the incident optical power so that the spectra can be compared. The vertical scale is approximately 12% of the peak intensity of the WL exciton.

Upon closer examination of Fig. 2a, one notices that some of the vertical emission lines contain fairly narrow (0.2 – 0.4 meV) resonances. Since this is precisely what one expects, it is tempting to assign each resonance to an excited state in the dot from which emission is collected. However, a plot of a few selected PLE spectra shows that the situation is not that simple, as resonances in more than one line frequently occur at the exact same excitation energy. (See Fig. 3). Since the spectra are uncorrelated outside their shared resonances, and since the energy spacing between the lines is essentially random, most of the lines in such a set must originate from separate quantum dots. These observations imply the existence of a mechanism for scattering carriers between different quantum dots.

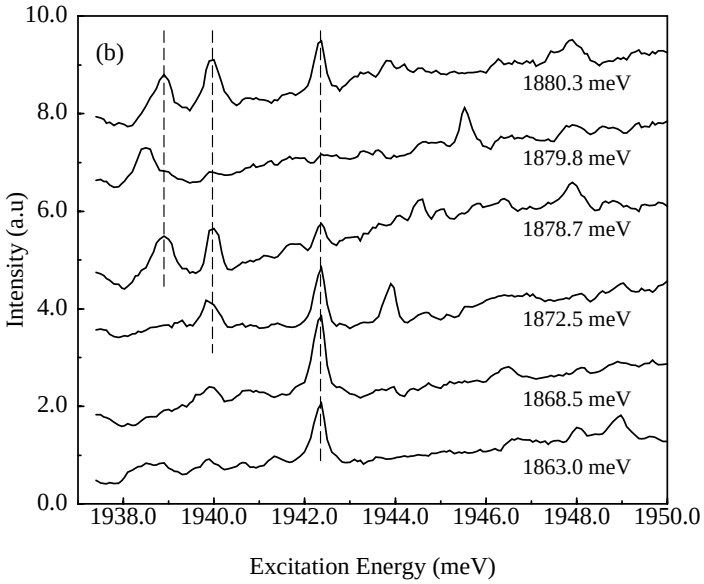


**Figure 3.**
Six simultaneously acquired PLE spectra of different quantum dots as indicated in Fig. 2a. The label of each trace is the detection energy. The dashed lines indicate resonances occurring simultaneously in more than one dot. The traces are offset vertically for clarity.

The simplest mechanism for the origin of the carrier scattering is direct dot-to-dot tunneling. Energy conservation requires such a process to be assisted by the emission or absorption of phonons, and a simple estimate [15] using reasonable parameters for the sample – 10 nm nearest-neighbor separation, and a 50 meV tunneling barrier – show it to be much slower than a typical exciton life time (~1 ns). Thermal depopulation of the dots is another possible candidate mechanism. It has, in fact, been previously invoked to explain similar phenomena in SADs [8]. In that case however, no dot-to-dot transfer was observed at 4 K, the temperature at which these experiments were performed. It is still possible that the high optical power density immediately below the near-field tip ($10^2$ – $10^3$ W/cm$^2$) raises the local electronic temperature significantly above the background. If that were the case, though, we would expect the frequency and magnitude of the scattering to be non-linear in the optical power. This is not the case [15], and we can therefore exclude this possibility as well. To explain our observations, we must therefore invoke the presence of the WL localized states discussed above. If such states are taken into account, carriers travelling from one quantum dot to another can do so through two or more tunneling events via those states. This provides a coherent picture of the SAD-WL system, explaining all our observations within a single framework.

Finally, one more phenomenon is visible in the PLE data, when the laser is tuned to wavelengths closer to the detection wavelengths. Data taken in this regime, exemplified by the plot in Fig. 4, show diagonal stripes of increased intensity across the data set in addition to the phenomena discussed above. Since the slope of these stripes equals 1 within the precision with which we can measure, we believe they are phonon lines, as has been observed in previous far-field measurements of the same sample [9]. We can discern at least a dozen different lines in the figure. This very rich phonon spectrum is not unexpected, since we are probing a volume containing complicated interfaces between two ternary semiconductor alloys. Each alloy will itself give rise to at

least two LO-modes from the bulk, each of which will engender several interface phonon modes at the boundaries between the barrier and dot materials.

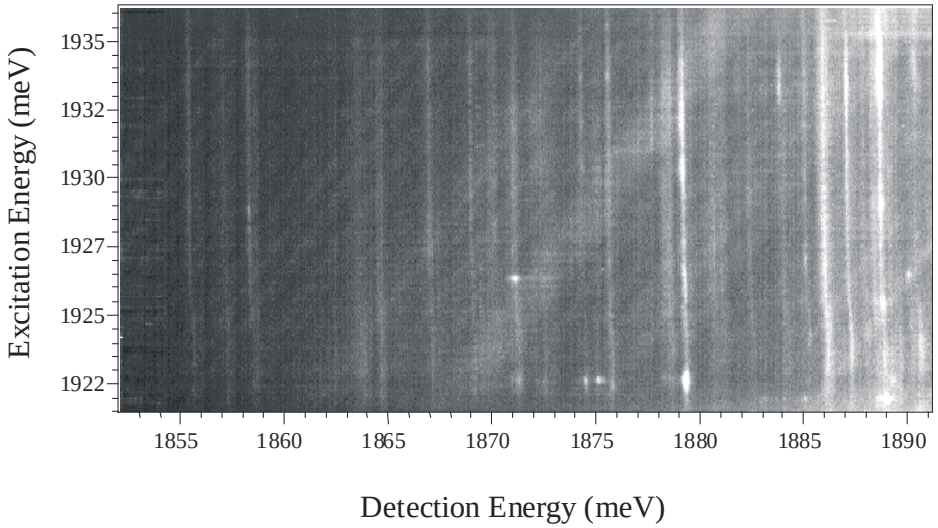


**Figure 4.** Plot of a PLE data set similar to Fig. 2a, but exciting closer to the detection energies. Phonon lines show up as diagonal stripes with slope 1.

## CONCLUSION

We can, with high confidence, conclude that we have observed scattering of carriers between individual quantum dots mediated by localized states in the WL. This effect is likely to be common in SADs, depending on the length scale and severity of the potential fluctuations creating the WL states. In general, the matrix surrounding the quantum dots is likely to exert a strong influence on the physics of SAD systems, a fact that should be kept in mind whenever treating the quantum dots as near zero-dimensional.

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