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Investigation of coupling in naturally occurring GaAs quantum dots

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Abstract

We report on the photoluminescence (PL) imaging of GaAs quantum dots (QDs) formed by islands of quantized thicknesses in a 2 nm thick $Al_{0.3}Ga_{0.7}As/GaAs/Al_{0.3}Ga_{0.7}As$ quantum well (QW) by means of near-field scanning optical microscopy. Discrete spectra associated with the QDs are observed and studied. Individual QDs are imaged with a resolution of 150 nm. Energy degeneracy which is sometimes observed between nearby QDs suggests the possibility of quantum mechanically coupled dots. Constraints on the observation of coupling are discussed.

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Semiconductor quantum dots (QDs) have generated a great amount of interest because of their novel physics and because of the recent trend toward ever smaller semiconductor devices. A particularly interesting potential application of QDs is in quantum information technology where they would serve as quantum bits (qbits). In this application QD states between different QDs must interact or entangle in a controlled way. There have been recent reports of coherent control of QDs [1] and QD entanglement [2,3]. A widely used and effective technique of probing single QDs is photoluminescence (PL) spectroscopy. In this way, the energy levels of QDs can be directly measured nondestructively. However, the high surface density of typical QD samples makes the resolution of single dots difficult to achieve. Traditional far-field microscopy can obtain resolution on the order of 400 nm while imaging with a solid immersion lens (SIL) extends this range down to about 250 nm [4,5]. Near-field methods, either *in situ* nanoapertures [6] or scanning probes [7], can achieve 50 nm resolution while sacrificing optical throughput. Unlike nanoapertures, near-field scanning optical microscopy (NSOM) has the capability to spatially scan a QD and its environment with the highest resolution presently possible.

Though a wide variety of QD types exist [8,9], in this paper we report on the so-called "natural" or "accidental" dots in GaAs/GaAlAs quantum wells (QWs). These QDs arise from monolayer fluctuations at the well interfaces of narrow QWs that lead to confinement in the growth plane [6,10]. We examine the possibility of observing naturally occurring coupled QDs using NSOM techniques. In the next section we describe the QD samples and NSOM experiment. This is followed by a presentation of the NSOM data and a discussion of the observations. We conclude with an analysis of a model of the naturally occurring QDs and the spatial and spectral resolution issues involved in studying such coupled QDs by NSOM techniques.

The QW sample used in this report consisted of a stack of ten GaAs QWs of varying thickness from 20 to 2 nm separated by 22.5 nm layers of $Al_{0.3}Ga_{0.7}As$ with a 5 nm GaAs cap layer. At 27.5 nm away, only the narrowest QW was close enough to the surface to be measured in the near field. To measure the PL from the QDs, the NSOM was operated in the so-called collection mode. In this mode,

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laser light was focused onto the coherent fiber bundle and collected with the NSOM tip along with PL from the sample. The collected light was then focused onto the entrance slit of a single-grating spectrometer. A relatively large NSOM probe diameter of 150 nm was typically used to get a sufficiently large throughput. The experimental scheme is illustrated in Fig. 1.

The primary reason for using collection mode over illumination mode is because of exciton diffusion. In illumination mode, where the sample is illuminated by the NSOM tip, above gap excitons can be created under the tip but then diffuse to a QD some distance away. Since diffusion lengths can be on the order of 250 nm or more [5], spatial resolution is lost. This is not a problem in collection mode since PL is collected only from excitons directly under the tip.

A typical spectrum taken at 4 K is shown in Fig. 2. The broad humps in the spectrum correspond to distributions of lateral sizes of monolayer fluctuations on backgrounds with thicknesses of 7, 6, and 5 monolayers. Clear signatures of QDs are seen as sharp features in the low energy range of the spectrum. The density of these lines is roughly 400 per μm^2. The energy of each line generally corresponds to the ground state energy of an exciton in each QD though some lines likely originate from excited states [7]. Sufficient spatial and spectral resolutions are needed to observe these lines. To demonstrate this, the average spectrum from a scan roughly 1 μm^2 in size is also shown in Fig. 2. In this average spectrum the sharp features are washed out.

Using a monolayer thickness of 0.2827 nm [11], the QDs are taken to be 8-monolayer thick islands on a background of a 7-monolayer thick QW. As the lateral size of the monolayer island increases the lateral confinement energy decreases, and the spectral signature is red shifted. An infinitely large island corresponds to a perfect 8-monolayer thick well. Following the interpretation of Wu et al. [5], the energy of a perfect 7-monolayer well corresponds to a node in the spectrum. The spectrum has a local minimum, indicated in Fig. 2 as E_7, at about 1748.5 meV. The spectrum similarly goes to zero at about $E_8 = 1725.5$ meV for 8 monolayers. The energy of a particular QD is blue shifted from E_8 by its lateral confinement energy, determined by the size of the monolayer island forming the QD.

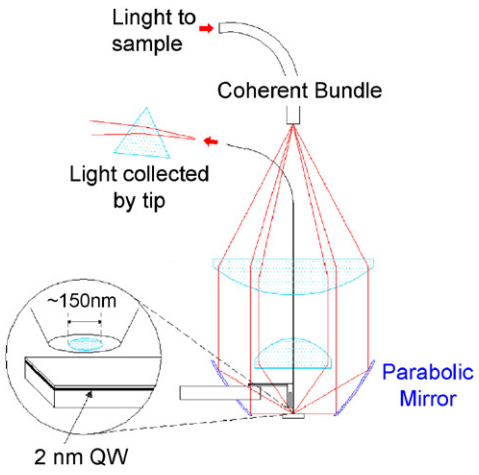

Fig. 1. Experimental setup for near-field photoluminescence of GaAs quantum dots.

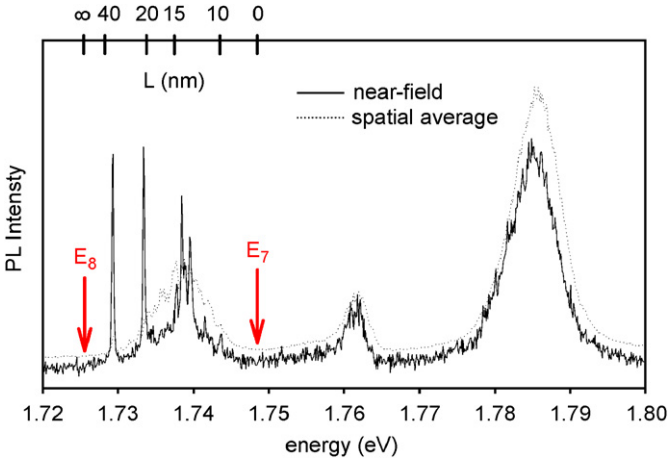

Fig. 2. Typical PL spectrum from light collected through the NSOM tip. E_x indicates the ground-state energy of an exciton in a perfect well x monolayers thick. The lateral size of a monolayer island L is plotted to illustrate its influence on the PL energy. The dotted line shows the average of spectra over a spatial scan of roughly 1 μm^2. The excitation energy is 1.96 eV.

The PL energy E_{QW} of a QW can be calculated in terms of the GaAs band gap E_G, the sum of the electron and hole confinement energies E_z, and the exciton binding energy E_B as

$$E_{QW} = E_G + E_z - E_B. \quad (1)$$

The electron and hole confinements are calculated separately as particles in a 1D square well potential. The height of the potential for the electron (hole) is just the conduction (valence) band offset of the semiconductors. The difference in mass m in GaAs and M in $Al_xGa_{1-x}As$, will introduce a discontinuity in the wavefunction at the interfaces [11]. This leads to an equation relating the energy E of a particle in a finite square potential of height V and dimension a which is

$$a = \frac{2\hbar}{\sqrt{2mE}} \tan^{-1}\sqrt{\left(\frac{m}{M}\right)\frac{V-E}{E}}. \quad (2)$$

The lowest energy of the even wavefunction solution is the ground state and is equal to the confinement energy. With appropriate values [12], Eqs. (1) and (2) can be used to predict the PL energies of the GaAs QWs. For a narrow QW with a thickness less than the 3D exciton Bohr diameter of about $2a_B = 22$ nm, the estimation of the exciton binding energy requires consideration of the nearly 2D motion [13]. Using Eqs. (1) and (2) with the 2D binding energy, E_7 and E_8 were calculated to be 1744.0 and 1721.8 meV, respectively, which are consistent with the observed energy spectrum.