Fine Structure Splitting in the Optical Spectra of Single GaAs Quantum Dots

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We report a photoluminescence study of excitons localized by interface fluctuations in a narrow GaAs/AlGaAs quantum well. This type of structure provides a valuable system for the optical study of quantum dots. By reducing the area of the sample studied down to the optical near-field regime, only a few dots are probed. With resonant excitation we measure the excited-state spectra of single quantum dots. Many of the spectral lines are linearly polarized with a fine structure splitting of 20–50 μeV. These optical properties are consistent with the characteristic asymmetry of the interface fluctuations.

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In this Letter we describe the polarization dependence of the optical spectra of single naturally formed GaAs quantum dots. Most previous optical studies of quantum dots (QDs) have probed large ensembles which have led to inhomogeneous broadening of the spectral features. However, recently several groups have shown that it is possible to study single QDs with photoluminescence (PL) either by reducing the size of the sample, [1] by cathodoluminescence [2,3], or by reducing the size of the laser spot on the sample through microscopic [4,5] or optical near-field techniques [6]. Here we use a similar technique whereby we combine high spatial and spectral resolution optics with excitation spectroscopy to study in detail the spectrum of a single QD [7]. With improved resolution we are able to resolve the spectral lines and to study the polarization dependence of the PL spectrum of an individual QD. We often find that the PL is linearly polarized along the (110) crystal axes and observe a fine structure splitting in each of the spectral lines. These results are analogous to the early days of atomic spectroscopy as improvements in techniques allowed the observation of fine structure splittings in the optical spectra. However, the physical phenomena responsible for the effects presented here are unique to the quantized condensed matter system.

The QDs we have studied were formed naturally by interface steps in narrow quantum wells [4–7]. Specifically, the electrons and holes become localized into QDs in regions of the quantum well that are a monolayer wider than the surrounding region and, therefore, have a slightly smaller confinement energy. These well width fluctuations arise from monolayer-high islands at the interfaces which are randomly formed on the growth-interrupted surface by the migration of the cations to step edges. By interrupting the growth these islands can grow to diameters larger than the exciton Bohr diameter (20 nm). A scanning tunneling microscope image of a growth-interrupted GaAs surface grown under similar conditions as our quantum dot sample is shown in Fig. 1. Large monolayer-high islands of varying lateral sizes are evident, and *the islands tend to be elongated along the [$\bar{1}$10] crystal axis.* Thus we intuitively expect that the optical properties associated with the localized excitons will reflect this characteristic interface structure. In fact, as we will show, this structural asymmetry can impose a linear polarization on the QD exciton and results in a fine structure splitting.

The sample studied consists of a GaAs buffer followed by 5 GaAs quantum wells with varying widths separated by 25 nm $Al_{0.3}Ga_{0.7}As$ barriers. The last barrier layer is followed by a 50 nm GaAs cap. Here we consider only a 2.8 nm quantum well that was grown with 2 min growth interrupts at each interface. Electron-beam lithography and metal liftoff were used to open a series of apertures in an opaque 100 nm thick Al film deposited on the surface [8]. The holes, ranging in size from 25 to 0.2 μm, were spaced sufficiently far apart to allow optical probing of a single hole. The PL was excited and detected through the same hole with either an argon laser or a Ti:sapphire laser with a power density of $\approx$10 W/cm^2. The PL was detected with a Dilor triple spectrometer and a charge coupled device detector. The resolution of the

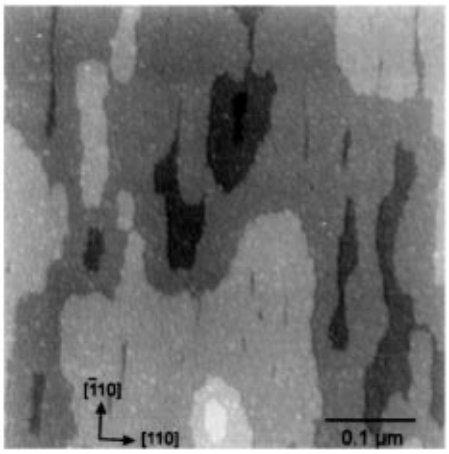

FIG. 1. A representative scanning tunneling microscope image of a GaAs surface which has been kept at growth temperature for several minutes under an As flux before being cooled and measured. The changes in gray level correspond to 1 monolayer-high changes in height. One observes that the islands are aligned approximately along the [$\bar{1}$10] axis.

PL spectroscopy was determined by the spectrometer and is 30 μeV. The resolution of the excitation spectroscopy was determined by the laser linewidth which is $\sim$7 μeV. The polarization was analyzed using a half wave plate and a polarization analyzer. We also note that we have measured the magnetic field dependence of the QD discussed in detail below and find that the measured linear polarization becomes circular at sufficiently high magnetic field. Therefore, anisotropies in the optical system cannot account for the observation of linear polarization of the QD discussed below.

The PL from this sample as measured through a 25 μm aperture is shown at the bottom of Fig. 2. The PL arises from the heavy hole exciton and is split into a doublet with a splitting of 11 meV. The components of this doublet are due to recombination of the exciton in 10 and 11 monolayer-wide regions of the quantum well. With decreasing aperture size (Fig. 2) the two broad peaks split into a series of extremely sharp lines, the number of which decreases with decreasing aperture size. Previous studies have shown that these lines arise from excitons localized into QDs by the interface fluctuations [4–7]. The intensities of the sharp lines are extremely sensitive to excitation energy. This behavior is shown in Fig. 3 for a 1.5 μm aperture. The color scale represents the intensity of the PL recorded as a function of both the PL and laser energy. For this plot the laser was stepped in units of 100 μeV. A horizontal line profile of this plot shows the PL spectrum at a particular laser energy while a vertical slice gives the excitation spectrum for a particular PL energy. The diagonal line at $E_{ex} = E_{PL}$ can be viewed as either the excitation energies for the PL spectra or the ground state energies for the excitation spectra. The bandpass of the spectrometer did not allow detection of the PL within 0.8 meV of the laser.

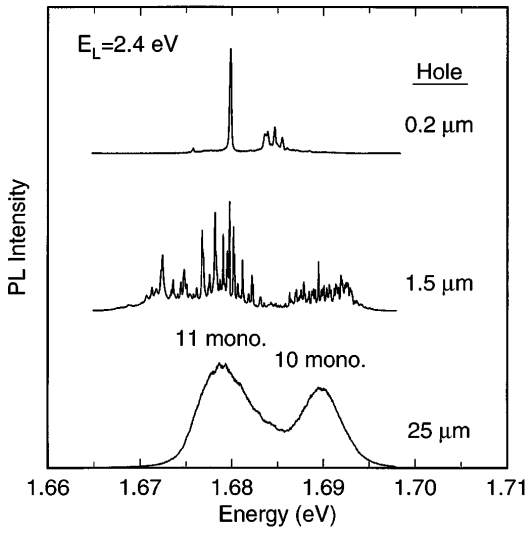

FIG. 2. Nonresonant PL spectra excited and detected through apertures with diameters listed.

In Fig. 3(a) qualitative differences in the excitation spectra for the 10 and 11 monolayer regions are evident. Within the 11 monolayer region there are a relatively small number of PL lines (about 15). The excitation spectra of most of these lines [Fig. 3(b)] contain discrete resonances which merge into quaisicontinua near the energy of the 10 monolayer region. Such discrete spectra are a signature for excited states of QDs. To estimate the typical size of the QDs we take the simplest model for the QD potential which is to assume that it is defined by a monolayer-high interface island. In the growth direction the potential offset is the $Al_{0.3}Ga_{0.7}As$ barrier (300 meV), while in the lateral dimensions the potential is defined by the difference in confinement energy due to the monolayer difference in well width (11 meV). From the energy spacings of the QDs, we estimate that a typical QD size is on the order of 100 nm. Therefore, the area of the sample from which these PL lines originate is about 10^4 nm^2 which is $\sim$2% of the area in the 1.5 μm aperture. In contrast, in the 10 monolayer region the PL lines are much more closely spaced, and, although there is sharp structure in the excitation spectra, the resonances are continuous and extend down to the detection energy. This quasicontinuous behavior indicates that the 10 monolayer regions are so closely spaced that they have merged into a 2D quasicontinuum.

Most of the QDs in the 11 monolayer region have unique excitation spectra. These QDs are assumed to be isolated and independent. This is not surprising because these QDs represent 2% of the total area of the quantum well and on average are well separated from other QDs with similar energies. There are some QDs which have more continuous excitation spectra with some common lines. We suspect that these QDs are coupled. It is easy to appreciate that such behavior can occur from an examination of the geometries of the islands shown in Fig. 1. We focus only on the behavior of isolated QDs. In particular, we discuss in detail the spectrum of the QD indicated by the arrows in Figs. 3(a) and 3(b).

The PL of the ground state (E_0) of this QD, along with the first four lines of its excitation spectrum, are plotted in Fig. 4. All of the lines are extremely narrow. These lines are well fit by Lorentzians, and the linewidths vary from 32 to 51 μeV (FWHM). Using the first transition energy of 2.5 meV ($E_1 - E_0$) and a square well potential of 11 meV, a lateral dimension of $\sim$40 nm is calculated.

An examination of Fig. 1 indicates that the islands are asymmetric with an overall elongated shape. This lack of symmetry lifts the degeneracy and polarizes the optical spectra. [9,10] The PL from the E_0 line was obtained by exciting and detecting with polarizations parallel to each other and to one of the (110) axes. With nonresonant excitation (i.e., 2.41 eV), the spectra are independent of the polarization of the source. However, the energies and intensities of the PL lines depend on the spectrometer polarization, shifting slightly in energy with small changes

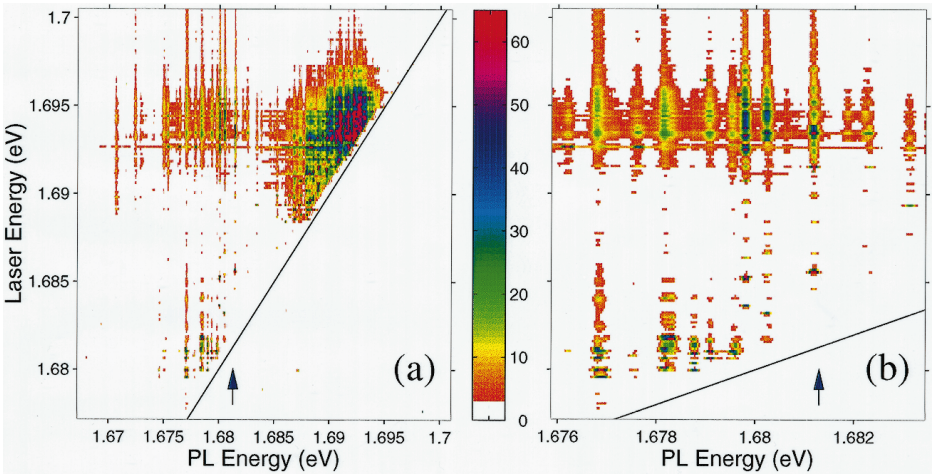

FIG. 3 (color). PL intensities are plotted according to the color scale as a function of PL energy and excitation energy. (a) Both the 10 and 11 monolayer regions, while (b) is an expanded view of the QD region in the 11 monolayer region. The arrow points to the QD discussed in the text and in Fig. 4.

in intensity. This behavior arises because the PL lines are composed of doublets with the two components linearly polarized along perpendicular directions. If the excitation spectrum is obtained by detecting either one of the two components, each of the excited states is also observed to be split into polarized doublets. The resulting excitation spectra are shown in the inset to Fig. 4 for the first and fourth excited states when excited and detected with the (x', x') and (y', y'), configurations, where $(*, *)$ corresponds to the polarization of the (incident, detected) light and x' and y' are along the two (110) axes. The magnitude and the sign of the splitting vary as listed in the table in Fig. 4. In the crossed polarization configurations the PL is much weaker, which indicates a polarization memory as the exciton relaxes from the excited to the ground state. If the polarizations are rotated to lie along the (100) axes, this polarization behavior is not observed. Specifically, doublets are measured in the excitation spectra with both parallel and crossed polarizations. While the PL intensities of the two components of the ground state are approximately equal ($<$20% difference), some of the excited states have intensities which differ considerably (Fig. 4). We also note that energy splittings with varying magnitudes were observed in many of the PL lines. We measured the excitation spectrum of four QDs within two separate apertures with high resolution and found energy splittings in the excited states of all four. Three of the four were linearly polarized along the (110) axes.

By measuring the PL from a larger (5 μm) aperture in which a larger number of QDs were excited, we obtain additional information on the relative number of PL lines which are linearly polarized. We find that a large fraction of those lines that are sharp and exhibit sharp resonances are linearly polarized along the (110) axes. However, the spectrum also contains a large amount of broader background emission which is not polarized. This unpolarized emission may originate from less localized or delocalized excitons.

We qualitatively consider the origin of the observed polarization dependence. The relative intensities of the different lines [e.g., the difference in intensities between the $(x'x')$ and $(y'y')$ polarizations for E_3, etc.] likely arises from mixing of the heavy and light holes due to the QD potential [11]. Inversion asymmetries in the QD potential and/or local uniaxial strain coupled with spin-orbit interaction may lead to a lifting of the spin degeneracy in the electron and hole single particle states [12]. Although calculations for the magnitude of the spin splittings have not been reported for QDs, spin splittings at finite wave vector have been observed for quantum well structures [13–15].

In addition to this splitting, the exciton states in QDs can also be split by the exchange interaction. In quantum wells the heavy hole exciton is split by exchange into a doubly degenerate level with total angular momentum $J = 1$ and a doublet with $J = 2$ [9,10]. Only the $J = 1$ states are optically active. Elongation of the in-plane QD confinement potential will lift the remaining degeneracy via the exchange interaction [9,10]. There is an intuitively simple way to understand this. It has been shown that the magnitude of the splitting induced by the long-range part of the exchange interaction is equivalent to modeling the system as a dielectric medium and calculating the electrostatic response of the system [16]. Although this calculation has not been reported for a QD, it has been performed for a quantum well [16,17]. In this case, when the polarization of the exciton is normal to the plane of the quantum well, the depolarization field associated with the quantum well interfaces leads to a restoring

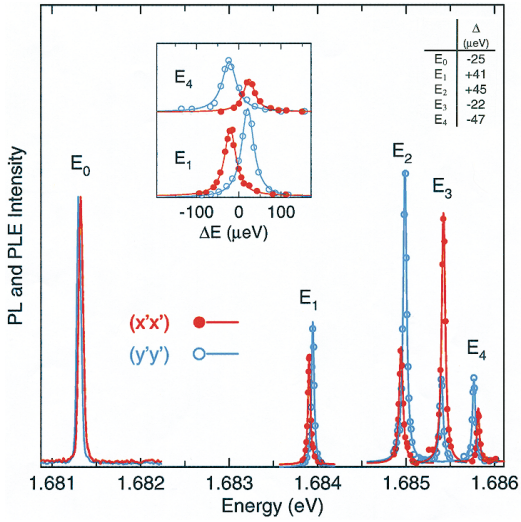

FIG. 4 (color). The spectrum of the QD indicated by the arrow in Fig. 3. E_0 is the ground state PL while E_1-E_4 are the first four excited states as measured by excitation spectroscopy in two polarization configurations. The inset shows an expanded view of the polarization dependence of two of the excited state lines, and the table lists the magnitude and sign of the splittings (Δ).

force which increases the energy of the exciton by about 1 meV. Because excitons polarized in the plane of the quantum well do not experience a depolarization field, there is a 1 meV splitting between the energies of excitons polarized parallel and perpendicular to the plane of the quantum well. In the QD case, depolarization fields are created for excitons polarized along the plane of the quantum well. For an elongated QD, the magnitude of the depolarization field will depend on the orientation of the polarization of the exciton with respect to the principal axes of the QD, leading to a fine structure splitting. The two components of the resulting doublet will be polarized along the principle axes of the QD potential. In our case the principle axes will tend to be along the (110) axes because the islands which define the QDs tend to be aligned this way (see Fig. 1). We also note that in this model the magnitude of the depolarization fields will vary not only with the dimension of the QD but also with the number of nodes in the exciton's wave function, as well as with how those nodes are aligned with respect to the polarization of the exciton. Therefore, the splittings in the excited states will vary in magnitude and sign. Because the quantum well width is much smaller than the in-plane dimensions of the QD, the depolarization fields for excitons polarized in the plane are expected to be small. We conclude that this model is in qualitative agreement with all of the experimental observations. The splitting, resulting either from the exchange interaction or from the spin-orbit interaction, will depend on the details of the Qdot potential considered—especially on the amount of asymmetry. However, in the sense that real QDs will almost certainly contain a certain amount of asymmetry, the fine structure splitting observed here is likely to be quite general.

In conclusion, with high spatial and spectral resolution we have obtained the spectrum of a single QD formed by the potential fluctuations of a narrow quantum well. The lines are extremely narrow with linear polarization and fine structure splitting. This behavior likely arises from the asymmetry of the interface fluctuations which define the QDs.

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[1] J.-Y. Marzin, J.-M. Gerard, A. Izrael, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994).
[2] R. Leon, P. M. Petroff, D. Leonard, and S. Fafard, Science **267**, 1966 (1995).
[3] M. Grundmann *et al.*, Phys. Rev. Lett. **74**, 4043 (1995).
[4] A. Zrenner, L. V. Butov, M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann, Phys. Rev. Lett. **72**, 3382 (1994).
[5] K. Brunner, G. Abstreiter, G. Böhm, G. Tränkle, and G. Weimann, Phys. Rev. Lett. **73**, 1138 (1994); Appl. Phys. Lett. **64**, 3320 (1994).
[6] H. F. Hess, E. Betzig, T. D. Harris, L. N. Pfeiffer, and K. W. West, Science **264**, 1740 (1994).
[7] D. Gammon, E. S. Snow, and D. S. Katzer, Appl. Phys. Lett. **67**, 2391 (1995).
[8] A general issue in optical near-field spectroscopy concerns the effect of the mask on the exciton itself. Although we cannot rule out all interactions, we believe that such effects cannot explain our polarization data. Specifically, the principal axes of the optical properties are along the (110) axes. In contrast, the 1.5 μm aperture was measured and found to have a slight ellipticity (diameters of 1.6 and 1.4 μm), but along the (100) axes.
[9] H. W. van Kesteren, E. C. Cosman, W. A. J. A. van der Poel, and C. T. Foxon, Phys. Rev. Lett. **61**, 129 (1988); Phys. Rev. B **41**, 5283 (1990).
[10] E. Blackwood, M. J. Snelling, R. T. Harley, S. R. Andrews, and C. T. B. Foxon, Phys. Rev. B **50**, 14 246 (1994).
[11] T. Tanaka, J. Singh, Y. Arakawa, and P. Bhattacharya, Appl. Phys. Lett. **62**, 756 (1993).
[12] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Editions de Physiques, Les Ulis, France, 1988).
[13] P. D. Dresselhaus, C. M. A. Papavassiliou, R. G. Wheeler, and R. N. Sacks, Phys. Rev. Lett. **68**, 106 (1992).
[14] B. Jusserand, D. Richards, H. Peric, and B. Etienne, Phys. Rev. Lett. **69**, 848 (1992); Phys. Rev. B **51**, 4707 (1995).
[15] P. V. Santos and M. Cardona, Phys. Rev. Lett. **72**, 432 (1994); Phys. Rev. B **51**, 5121 (1995).
[16] L. C. Andreani and F. Bassani, Phys. Rev. B **41**, 7536 (1990).
[17] M. Nakayama, Solid State Commun. **55**, 1053 (1985).