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Magneto-optical studies of self-organized InAs/GaAs quantum dots

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Magneto-photoluminescence and photoluminescence excitation (PLE) measurements of self-organized InAs/GaAs quantum dots are reported. For fields applied along the growth direction the first excited-state transition exhibits a linear Zeeman splitting, consistent with a transition between the ground electron state and an excited p-like hole state $(\Delta n \neq 0)$. The hole mass determined from the splitting is in good agreement with the value obtained from a $\mathbf{k}\cdot\mathbf{p}$ calculation for the highly strained dots. The size of the splitting decreases as the field is rotated away from the growth direction, demonstrating the highly anisotropic nature of the carrier wave function. A comparison of spectra recorded at high dot carrier occupancies ($\gg 1$) in photoluminescence and at low carrier occupancies ($\ll 1$) in PLE reveals the influence of many-carrier Coulomb interactions. [S0163-1829(98)50304-9]

Self-organized quantum dots are of considerable contemporary interest as they provide zero-dimensionality (0D) systems with both large energy-level spacings and high optical quality.[1] Of particular interest is the form of the electronic states and resultant optical transitions,[2,3] the values of carrier effective masses[4] which are expected to be strongly modified by the effects of both strain and confinement, and the nature of carrier relaxation mechanisms.[1,5] In addition, excitonic effects in 0D are expected to result in a strong modification of the single-particle spectra,[6] with further modifications occurring as additional carriers are added.[7,8] In the present paper, a magneto-optical study of self-organized InAs/GaAs quantum dots is reported. The symmetry and anisotropy of the dot electronic wave functions are determined and the nature of the corresponding optical transitions deduced. The in-plane hole mass is measured and compared with $\mathbf{k}\cdot\mathbf{p}$ calculations of the strain and confinement modified carrier effective masses. A comparison of optical spectra recorded for both low and high dot carrier occupancies allows the effects of many-carrier Coulomb interactions to be studied.

Self-organized InAs quantum dots were grown on (001) GaAs by source solid molecular-beam epitaxy. The details of the growth are described in detail elsewhere[1] and resulted, after the deposition of a nominal 2.4 ML of InAs, in square shaped dots of base length $\approx$12 nm, height $\approx$2 nm, and density $\sim 5\times10^{10}$ cm^{-2}, as evidenced by transmission electron microscopy (TEM).[9] The dots were overgrown by a 100-nm GaAs layer. Optical spectra were excited with either an Ar^+ laser [photoluminescence (PL)] or an Ar^+ pumped Ti:sapphire laser [photoluminescence excitation (PLE)]. The resultant luminescence was dispersed by a double-grating spectrometer and detected with a liquid-nitrogen cooled Ge p-i-n photodiode. Magnetic fields up to 14 T were applied by a vertical coil, optical access superconducting magnet. The experiments probe a large number ($\sim 10^6$) of dots.

The inset to Fig. 1 shows a high ($\sim$20 kW cm^{-2}) incident power-density Ar^+ PL spectrum. Under these conditions the carrier population of at least some of the dots is sufficient to allow the observation of two higher-energy, excited-state transitions T_2 and T_3, in addition to the ground-state transition T_1. T_2 and T_3 are separated from T_1 by 74 and 120 meV, respectively. The observation of the higher-energy T_3 transition implies a minimum hole occupancy of 6 for at least some of the dots ($2x$ ground s state + $4x$ excited p state). Many-carrier Coulomb interactions are therefore expected to be very important in the PL spectra, in contrast to

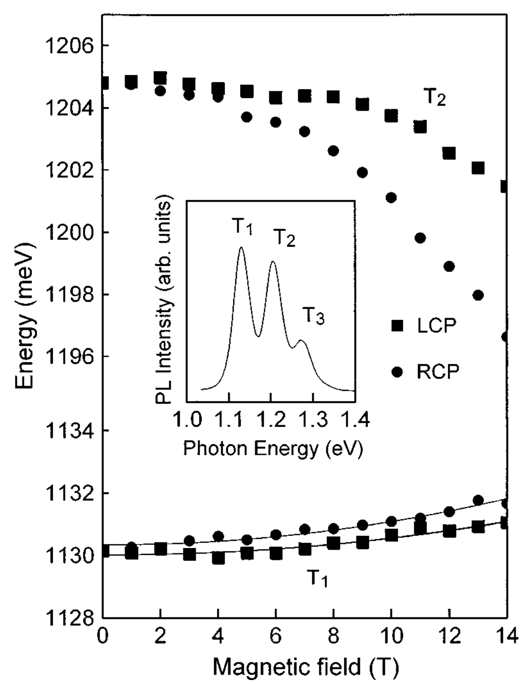

FIG. 1. High incident laser power PL transition energies plotted against the magnetic field. The field is applied along the growth axis and for each field, data points are plotted for right- and left-circularly polarized emitted light. The inset shows the zero-field spectrum. Note the break of scale between 1134 and 1196 meV.

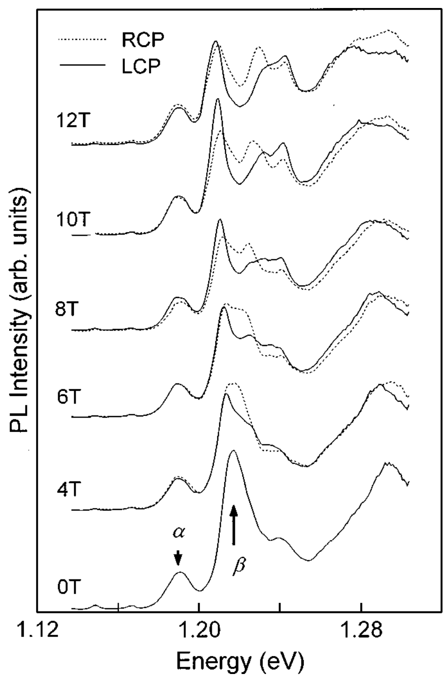

FIG. 2. PLE spectra, recorded for detection at the peak of the ground-state PL, as a function of the magnetic field applied parallel to the growth axis. For each field spectra recorded for right- and left-circularly polarized incident light are shown.

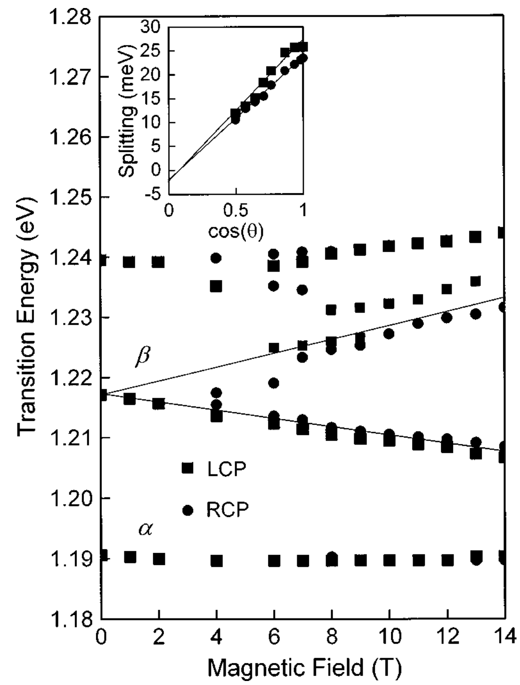

FIG. 3. PLE measured transition energies plotted against the magnetic field. The square and round symbols correspond to the different polarizations of the incident light. The solid lines, which are guides to the eye, show the large scale Zeeman splitting of feature β. The inset shows the size of the 14-T Zeeman splitting plotted against $\cos(\theta)$, where θ is the angle between the surface normal and the field direction.

PLE spectra, which are recorded for a dot occupancy of only one electron-hole pair (see below). The main part of Fig. 1 shows the magnetic-field dependence of the transitions T_1 and T_2 for the field applied along the growth axis. For each field, data points are plotted corresponding to analysis of the emitted light into left- and right-circularly polarized (LCP, RCP) components

T_1 exhibits a quadratic field dependence, the solid lines being fits to the experimental data. Coefficients of 7.6×10^{-3} and 5.5×10^{-3} meV/T^2 for the RCP and LCP components, respectively, are obtained. For free carriers the diamagnetic shift of the ground-state transition is given by[10,11]

$$\Delta E = \gamma e^2 B^2 \langle x^2 \rangle / 2\mu^*, \tag{1}$$

where μ^* is the electron-hole reduced mass and γ is a geometrical factor (≈ 0.5). Equation (1) thus permits the spatial extent of the wave function ($\sim 2\sqrt{\langle x^2 \rangle}$) to be deduced.[11–13] Electron and hole masses determined below give $\mu^* = 0.035 m_0$ and, with the assumption that $\langle x^2 \rangle$ has approximately the same value for electrons and holes, $2\sqrt{\langle x^2 \rangle} \approx 50$ Å is obtained from Eq. (1) and the average of the LCP and RCP experimentally measured diamagnetic coefficients. This result, which is in agreement with a previously reported value of 60 Å,[11] represents a lower limit for the spatial extent of the in-plane wave function as excitonic interactions result in a smaller diamagnetic shift compared to the free-carrier value given by Eq. (1).[10]

Figure 2 shows the effect of magnetic field on the PLE spectra for both LCP and RCP incident light. In PLE measurements on 0D systems the ground-state transition is not observed as the absorbed and emitted photons have the same energy (zero Stokes shift).[1] The resultant PL is hence obscured by the much stronger elastically scattered laser light. In addition, because in PLE the carriers are directly excited into the dots, their small absorption coefficient results in a very low carrier occupancy (~ 0.1). This is in strong contrast to the PL measurements described above, where the observation of the second excited transition implies an occupancy $\gtrsim 6$. The zero-field PLE spectrum in Fig. 2 contains a number of features, the two lowest energy ones being labeled α and β. Within the accuracy of the experiments the lower-energy feature α is unaffected by the magnetic field while the higher-energy feature β is split into a number of components. This behavior is more clearly seen in Fig. 3, where the peak energies are plotted against the magnetic field.

The magnetic-field behavior of β exhibits two different magnitudes of splitting, a large splitting (≈ 25 meV at 14 T), indicated by the solid lines in Fig. 3 which are guides to the eye, and a much smaller splitting (≈ 2 meV at 14 T) for the lower-energy component) between the different polarization configurations. Dots with cylindrical symmetry and a harmonic confinement potential have a doubly degenerate (neglecting spin) p-like ($m_L = \pm 1$) first excited state. In a single particle treatment such a state exhibits a linear, symmetrical Zeeman splitting for fields such that the magnetic energy is small compared to the confinement energy.[10,14] Although the present dots have a square shape, the zero-field degeneracy of the p-like states is preserved and a similar behavior is expected.[15] The approximately symmetrical large scale splitting of feature β is hence consistent with the expected Zeeman behavior of an excited p-like state.

This observation allows an effective mass for the carriers involved in the transition to be determined. The Zeeman splitting is given by $\Delta E = e\hbar B m_L / 2\mu^*$, where $m_L = \pm 1$ for

p states and μ^* is the reduced mass.[10] The measured splitting of 25 meV at 14 T allows a value for μ^* of $0.065m_0$ to be deduced. In order to interpret this value for μ^*, the expected electron and hole masses for the quantum dots are first required. The bulk InAs electron effective mass is $0.024m_0$, but both strain and confinement will considerably enhance this value. The strain enhancement can be determined by applying a three band $\mathbf{k}\cdot\mathbf{p}$ model[16] in which the bulk electron mass, fundamental band gap, and spin split-off band gap are used to determine the interband matrix element $\mathbf{P}^2$. This matrix element is then used with the strain modified band gap to calculate the band-edge mass in the strained material. Finally, nonparabolicity is added using the model of Gauer *et al.*[17] and confinement energies from the calculations of Cusack, Briddon, and Jaros.[17] This treatment results in an electron effective mass of $\approx 0.07m_0$. The in-plane heavy-hole mass in the limit of totally decoupled light and heavy-hole bands, which should be a good approximation in the present highly strained system, is determined in a similar way. The strain modified light-hole mass for motion along the growth axis is first calculated using the three band $\mathbf{k}\cdot\mathbf{p}$ model. This is then used with the heavy-hole mass, which is assumed to be strain independent, to determine the modified Luttinger parameters γ_1 and γ_2.[18] These parameters are then used to calculate the in-plane masses. An in-plane heavy-hole mass of $\approx 0.07m_0$ is obtained, similar to the electron mass.

Calculations predict only one confined electron state but a number of hole states,[2,3] suggesting that the experimentally determined mass ($0.065m_0$) represents the in-plane hole mass. Our measurements exclude the possibility that the first excited transition involves both excited electron and hole states,[19] as the reduced mass for such a transition ($\approx 0.5 \times 0.07m_0$) would result in a Zeeman splitting approximately twice as large as that measured experimentally. Although for dots with cylindrical symmetry only interband transitions for which there is no change in orbital angular momentum are strongly allowed,[20] this selection rule is relaxed in square shaped dots, and transitions between electron and hole levels of different indices ($\Delta n \neq 0$) are predicted to be allowed,[3] consistent with the present conclusions.

The magnitude of the Zeeman splitting of β (see inset of Fig. 3) decreases as the angle (θ) between the surface normal and the field direction is increased, following a $\cos(\theta)$ dependence and, within the experimental accuracy, falling to zero for $\theta = 90°$ (in-plane field). This behavior, in which only the field component along the growth direction determines the Zeeman splitting,[10] is consistent with highly anisotropic dots whose spatial extent along the growth direction (~20 Å from TEM) is much smaller than their in-plane spatial extent and which is also much smaller than the minimum attainable magnetic length [$l_C = (\hbar/eB)^{1/2} \approx 70$ Å at 14 T]. The smaller scale splitting observed in the PLE spectra is attributed to spin splitting.[8] For the lower-energy branch the splitting of 1.9 meV at 14 T corresponds to an effective g factor of 2.3. The ground-state transition as measured in PL exhibits a splitting of 0.6 meV at 14 T, giving an effective g factor of 0.7.

Although β and T_2 exhibit different zero-field and magnetic-field behavior, it is very likely that they have a common origin, with the differences resulting from many-carrier Coulomb interactions in the PL emission process. β and T_2 have similar, although slightly different, zero-field energies. In PLE, β occurs 84 meV above the ground state (as determined by the detection energy), a value larger than the PL measured T_1-T_2 splitting of 74 meV. This difference can be explained by a many-carrier renormalization of the energy levels in highly occupied dots, the conditions for the PL measurements. Calculations predict a renormalization of ≈ -15 meV for both the ground- and first-excited-state transition of $Ga_{0.5}In_{0.5}As$ self-organized dots,[21] resulting in an approximate carrier density independent energy separation of the first two transitions within a common dot. However, this behavior is not expected to be reflected in the PL spectra. While the simultaneous excitation of a large number of dots results in a well-defined global average carrier occupancy, the occupancy of a given dot has a random value.[22] The total emission spectrum hence consists of the weighted average of the emission spectra corresponding to all possible dot occupancies.[21,22] In the present measurements T_2 is observed before the intensity of the ground-state transition T_1 saturates, indicating that T_1 is likely to arise from dots containing only one or two holes. In contrast, T_2 must originate from dots containing a minimum of at least three holes with the weaker[21] ground state, renormalized emission from these dots being obscured by the more intense recombination from the lower occupied dots (T_1). The difference between T_1 and T_2 therefore represents the underlying state separation less the renormalization of *only* the first-excited-state transition, consistent with a smaller value than that measured for β in PLE.

T_2, as observed in PL (see Fig. 1), exhibits an initial nonlinear decrease in energy with increasing field. At high fields, particularly for the RCP component, this decrease becomes linear, with a gradient (≈ 1 meV/T) equal to approximately half the PLE determined Zeeman splitting ($\approx 0.5 \times 1.9$ meV/T) of β (Fig. 3). This initial nonlinear field dependence of T_2 results from many-body interactions, as demonstrated by calculations of the emission spectra of self-organized dots containing three or more excitons.[21] The differences in both the zero-field energies of β and T_2 and their magnetic-field behavior is hence consistent with the influence of many-body Coulomb effects in the PL process, implying that both β and T_2 represent the same, first-excited-state transition. The reason for the absence of the higher-energy $m_L = +1$ transition in PL is unclear but may indicate rapid carrier relaxation to the lower-energy $m_L = -1$ state or the influence of many-body effects that can result in an asymmetry of the $m_L = \pm 1$ emission intensities.[21]

Finally we note that the behavior of features α and β in zero-field PL spectra, where they show a constant energy separation from the detection energy and an approximate coincidence of these energies [58(2×29) and 84(3×28) meV for α and β, respectively] with integer multiples of the InAs LO-phonon energy ($\hbar\omega_{LO} = 29.9$ meV), has led to proposals that these features are not purely electronic in origin but represent a carrier relaxation process involving the emission of multiple LO phonons ($\alpha \equiv 2$LO, $\beta \equiv 3$LO).[1,4,23,24] The model for this mechanism is described in detail elsewhere[1,5] but in its basic form it is difficult to reconcile with the very different magnetic-field behavior of α and β described in the present paper. At present it is not clear whether α and β are purely electronic in origin or involve a phonon component, which, at zero field, selects from an inhomogeneous distribution those transitions of energy equal to integer multiples of the LO-phonon energy. One possible explanation is that the nonradiative processes required by the zero-field model[1,5] may become suppressed in magnetic field by the field-induced shrinkage of the carrier wave functions and resultant increased carrier confinement within the dots. In the magnetic field the PLE process would hence no longer be dominated by carrier relaxation and instead would reflect the absorption into the electronic states of the dots.

In conclusion, magneto-optical measurements of self-organized InAs/GaAs quantum dots have permitted the determination of the symmetry and spatial anisotropy of the carrier wave functions. In addition, the nature ($\Delta n \neq 0$) of the first excited optical transition has been deduced and the in-plane hole mass has been determined. A comparison of PL spectra obtained for carrier occupancies $\gg 1$ and PLE spectra recorded for occupancies $\ll 1$ demonstrates the influence of many-carrier Coulomb interactions in the former process.

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