Electron-Hole Transitions between States with Nonzero Angular Momenta in the Magnetoluminescence of Quantum Dots

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Photoluminescence studies on InGaAs/GaAs quantum dots with diameters down to 20 nm have been performed in magnetic fields up to $B = 12$ T and have been compared to detailed calculations. The eigenstates of these cylindrical dots are characterized by a radial quantum number and an azimuthal quantum number corresponding to the orbital angular momentum. From the distinctly different magnetic field dependences of the several transitions we identify electron-hole recombinations involving states with 0, ±1, and -2 values of the angular momentum.

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In recent years there has been much interest in artificially structured low dimensional semiconductor systems such as quantum wires and quantum dots from the conceptual point of view and also for possible applications. Until recently, the work on low dimensional structures has focused mainly on quantum wires. From optical studies, such features as quantization induced changes of the band gap and the formation of higher lateral subbands have been reported for them [1–8].

Compared to the case of quantum wires, there have been relatively few quantitative studies on quantum dot structures. Such systems are of great interest because they permit one to observe a fabrication controlled transition from an infinite semiconductor crystal to artificial atoms. Previously there has been experimental and theoretical work on arrays of dots, particularly on their collective properties, such as magneto-optical studies of infrared absorption of magnetoplasmons in fairly large dot structures [9,10]. To date, however, relatively little work has been reported on the optical properties of quantum dots of sufficiently small sizes to approach the quantum limit [11].

In the present Letter we report on magnetoluminescence studies of InGaAs/GaAs modulated barrier quantum dots for magnetic fields normal to the quantum disk. In addition to the vertical quantum well confinement, the electron and hole states in the dots are characterized by a radial quantum number and an angular momentum quantum number. By comparing the experimental data with the magnetic field dependence obtained from detailed calculations, we are able for the first time to identify quantum dot transitions between electron and hole states with orbital angular momenta of 0, ±1, and -2. These states go over into two different Landau levels at high fields.

The InGaAs/GaAs dot patterns used here were fabricated by high resolution electron beam lithography followed by the removal of the top GaAs layer in the lateral barrier section be selective wet etching. A scanning electron microscopy (SEM) micrograph of dots with a diameter of about 30 nm is shown in Fig. 1. In these modulated barrier structures the carriers are localized in the InGaAs quantum well in the region where the GaAs top layer has not been removed by virtue of the smaller carrier quantization energy there [3]. The InGaAs/GaAs material system and the quantum well thickness were chosen to provide a particularly simple band structure. For quantum well widths of 5 nm and the indium content of 13% used here, there is only one confined electron state and one confined heavy hole state in the quantum well. In addition, the strain due to the lattice mismatch to the GaAs substrate causes a heavy hole–light hole splitting of about 60 meV in the samples. Thus hole band mixing is not significant in these structures.

Photoluminescence measurements were performed at a temperature of 2 K in an optical split coil magnetocryostat up to $B = 10.5$ T and in a cryostat with a solenoid up to $B = 12$ T. The magnetic field was aligned parallel to the growth axis of the structures. In this geometry the lateral confinement of the carriers is enhanced by the field. A cw Ar^+ laser (514.5 nm) was used for the excitation with power densities varying from about 15 W cm^{-2} to 15 kW cm^{-2}. To excite the dot pattern homogeneously the laser was focused down to a spot

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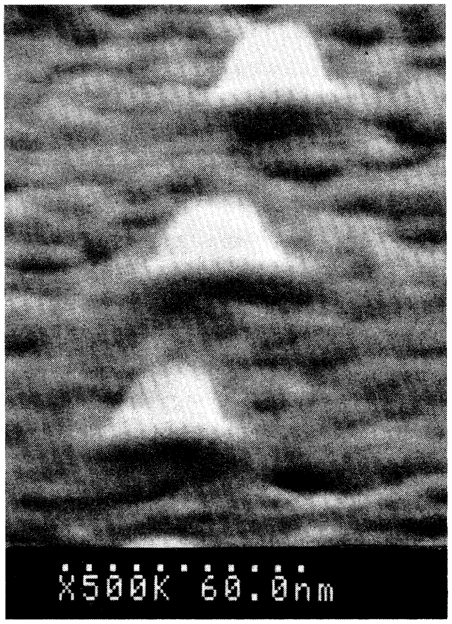

FIG. 1. Scanning electron micrograph of 30 nm $In_{0.13}Ga_{0.87}As/GaAs$ quantum dots.

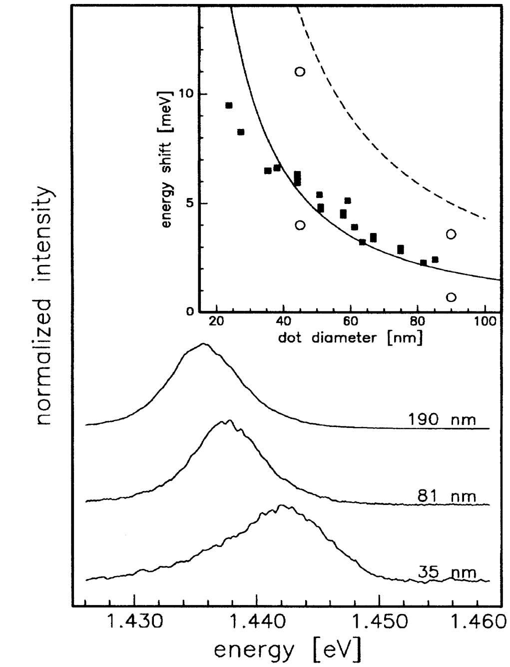

FIG. 2. Photoluminescence spectra of quantum dots with diameters of 35, 81, and 190 nm. The inset shows the measured shifts of the lowest transitions in photoluminescence as functions of dot size. The solid and dashed curves give the results of calculations described in the text. The open circles denote data obtained by photoluminescence excitation spectroscopy.

of about 150 μm in diameter, slightly larger than the dot array. The emitted light was dispersed by a 0.32 m single grating monochromator and detected by a liquid-N_2 cooled Si charge coupled device camera.

Figure 2 shows luminescence spectra for several diameters of $In_{0.13}Ga_{0.87}As/GaAs$ quantum dots taken at low optical excitation for zero magnetic field. The diameter of the GaAs cap layer was measured by high resolution SEM with an accuracy of about 5 nm. The SEM micrographs show that the dots are to a good approximation cylindrical. For decreasing dot diameters we observe a strong shift of the emission lines to higher energies. These shifts are plotted as a function of the dot diameter in the inset of Fig. 2. Data for 45 nm dots and 90 nm dots at $B = 0$ obtained in photoluminescence excitation spectroscopy are also shown. From the broadening of the spectral line with decreasing dot diameter we can extract a value for the size fluctuations of about ± 3 nm. These fluctuations are most likely due to local changes of the exposure conditions of the quantum dots due to the proximity effect. The lateral quantization energy in the smallest dots reaches values of up to 15 meV.

We made detailed numerical calculations for the single particle electron and hole states of these dots including the full lateral and vertical confining potentials. The angular momentum around the cylindrical axis is a good quantum number [12]. Schrödinger's equation in the remaining two coordinates (the growth direction z and the radial direction r) is nonseparable, and the calculations for them are performed by discretizing the equation and solving it using an iterative matrix diagonalization technique similar to that used by us earlier to treat quantum wires [7,8]. The states are then characterized by quantum numbers n_z, n_r, and the angular momentum m, which correspond to the number of nodes in each of these directions.

The calculated electron-hole transition energies at $B = 0$ of the two lowest lying states of these quantum dots are shown by the solid and dashed lines in the inset of Fig. 2. For dots smaller than 30 nm there is only one confined electron state and one confined hole state in the shallow lateral potential of these barrier modulated structures. The experimental data for these transitions are seen to be in agreement with the calculations with no adjustable parameters [13], which indicates that these structures are of high quality.

In order to observe emission from higher quantum dot states, high optical excitation was used. Figures 3(a)–3(c) show typical spectra at varying magnetic fields for 34, 35, and 41 nm diameter $In_{0.13}Ga_{0.87}As/GaAs$ quantum dots. Up to 8 electron-hole pairs per dot are excited by using excitation powers of up to 15 kW cm^{-2}. We discuss here the spectra of the 35 nm dots in detail, because for them the maximum number of different transitions could be observed. The higher transitions could be seen

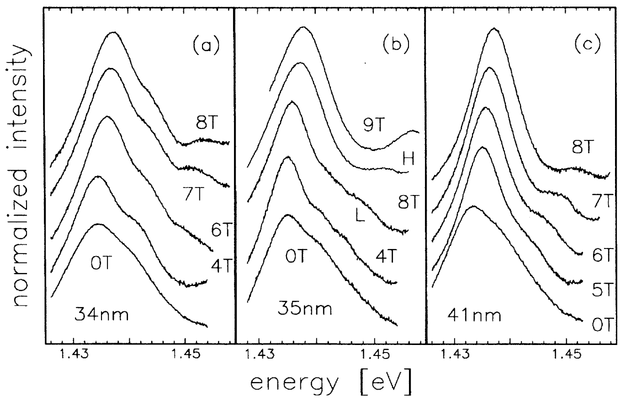

FIG. 3. High excitation photoluminescence spectra at varying magnetic fields for dots of 34 nm diameter (a), for dots of 35 nm diameter (b), and for dots of 41 nm diameter (c). The laser excitation power was 7.5 kW cm^{-2} except for the two topmost spectra of the 35 nm dots at $B = 8$ T (H) and at $B = 9$ T, where an excitation power of 15 kW cm^{-2} was used.

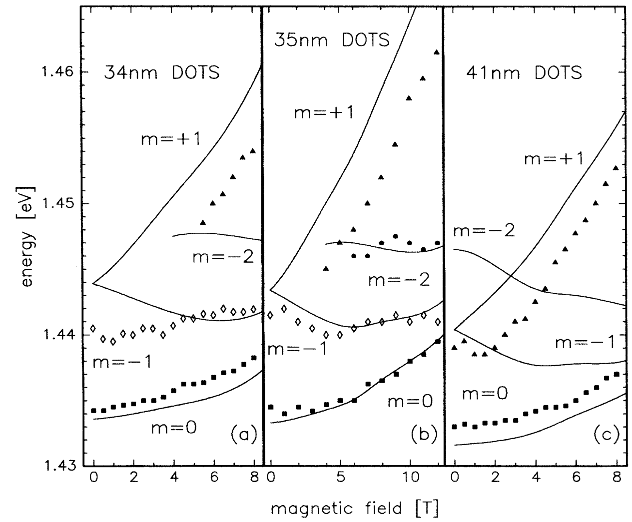

FIG. 4. Magnetophotoluminescence results for the ground state and higher lying transitions as functions of the magnetic field for modulated barrier dots with diameters of 34 nm (a), 35 nm (b), and 41 nm (c). The solid lines give the results of the calculations.

less well in the other samples because of the greater inhomogeneous broadening in them [14].

For the 35 nm dot sample at $B = 0$, a main line at $E = 1.435$ eV and a shoulder at 1.440 eV appear, and both of them show a weak dependence on B with increasing magnetic field. At $B = 4$ T, an additional very weak feature appears near 1.445 eV and also shifts only weakly with increasing B. Two different spectra at $B = 8$ T for different excitation powers are shown in Fig. 3 for the 35 nm dot sample. For lower excitation (L) the three features that were already observable at $B = 4$ T can again be seen. For increasing excitation power (H) these three features smear out to one spectral line centered around $E = 1.4375$ eV. In addition, a pronounced shoulder appears at 1.454 eV, and it shifts rapidly to higher energies with increasing field as can be seen from the high excitation spectrum at $B = 9$ T, where its energy is 1.456 eV.

Figure 4 shows the experimental data for the magnetic field dependence of the several transitions for the different dots compared to the results of our calculations. We are able to observe the magnetic field dependence of three, four, and two transitions for dots of 34, 35, and 41 nm diameters, respectively. In general, we can distinguish between two classes of magnetic field dependences. The low energy transition (squares in Fig. 3) and the second transition (diamonds in Fig. 3) depend rather weakly on the magnetic field, and they are seen to tend to converge for high magnetic fields. In addition, a transition observed at higher energy in the 35 nm dots depends only weakly on B [circles in Fig. 4(b)]. In contrast, the highest energy transitions (triangles) in all three samples increase rapidly with increasing field.

From our numerical calculations we find that there are three bound electron and hole states in the modulated barrier dots with diameters about 40 nm at zero magnetic field. The values for the quantum numbers n_z and n_r are those of the ground state, and the angular momentum states are given by $m = 0, \pm 1$. At zero field the states with $m = -1$ and $+1$ are degenerate, and $m = 0$ is the ground state. At nonzero magnetic fields the states with $m = \pm 1$ are split due to the interaction of the magnetic moment arising from the orbital angular momentum with the magnetic field which is given by $\frac{1}{2}\hbar\omega_c m$. For an arbitrary axially symmetric Hamiltonian the magnetic field dependence of the energy of a carrier can be written as the sum of this interaction energy, which is linear in B, plus a contribution which depends only on the magnitude of m and the magnitude of B [15]. Thus in the present case the difference in energy between the $m = +1$ and -1 states is $2\hbar(eB/\mu)$, where μ is the reduced mass. Their energy difference therefore should increase linearly with B, which is confirmed by the present experiments. In addition, we find that although the states corresponding to $m = -2$ transitions are not localized in the dot at $B = 0$, they are localized there for nonzero fields.

The strongest transitions are those for which the electron and hole angular momenta are the same. The calculated transition energies between these bound states are shown by the solid lines in Fig. 4. From the theoretical results for sufficiently high magnetic fields the lowest three states ($m = 0$, -1, and -2) go over into the lowest Landau level associated with a quantum well in a perpendicular magnetic field, and the $m = +1$ state is expected to go over into the next higher Landau level of the quantum well at high field. These trends are seen clearly in the data in Fig. 4.

The remarkably similar magnetic field behavior of the experimental data and the calculated transition energies allows us to identify the experimental transitions as being from electron and hole states having angular momentum quantum numbers $m_e = m_h = m$ equal to 0, ± 1, and -2 as indicated in Fig. 4. The calculated transition energies in Fig. 4 differ from the experimental values by a few meV. We attribute these differences to the high optical excitation used to observe the higher lying levels in these experiments which may result in many-body interactions between the carriers, which are not included here [16]. However, the key dependence of the transition energies on the magnetic field and on the angular momentum quantum numbers are described well by the present single particle calculations.

In summary, we have reported magnetoluminescence studies and numerical calculations for modulated barrier quantum dots with varying diameters which provide a rich picture of the carrier states of these systems. In particular, we have observed for the first time electron-hole transitions between states having nonzero orbital angular momenta.

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[11] For the InGaAs dots studied here with diameters of about 30–40 nm, the splitting between the ground state and the first excited state is on the order of 5 meV. For carrier temperatures of about 10 K and at low excitation intensities only the ground state in the dots is occupied by carriers (on average there is ≤ 1 electron-hole pair per dot). In the high excitation experiments discussed in the following, up to 8 electron-hole pairs are created in a dot. We find from experiments at widely varying laser powers that the influence of the excitation of several electron hole pairs on the transition energies is small (≤ 1 meV if the excitation power is increased from 0.5 to 15 kW cm^{-2}).

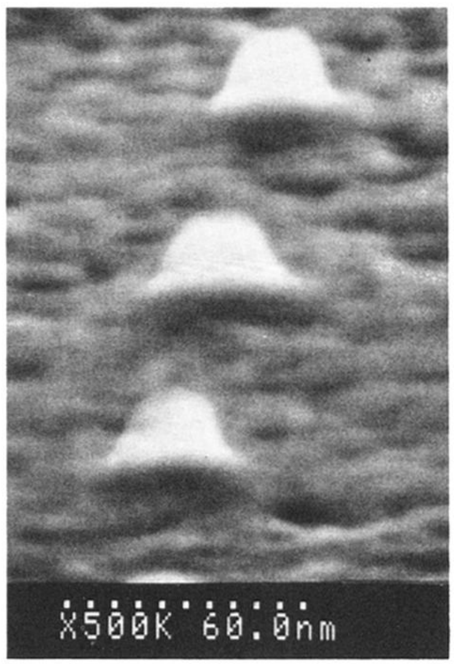
[12] The influence of a magnetic field on the eigenenergies of a harmonic oscillator has been trated analytically by V. Fock, Z. Phys. **47**, 446 (1928). The lateral and vertical confining potentials in the realistic dots in our experiments, unlike those of a harmonic oscillator, are abrupt and rather shallow, and numerical solutions are required for them.

[13] The same values of the electron and hole effective masses and band offsets which were used in the $B = 0$ work in Ref. [7] to describe modulated barrier InGaAs/GaAs quantum wires are used for the present calculations for quantum dots.

[14] For example, the photoluminescence spectra of the 34 nm dots in Fig. 3 show linewidths that are about 2 meV larger than the linewidths of the 35 nm dots.

[15] R.H. Dicke and J.P. Wittke, *Introduction to Quantum Mechanics* (Addison-Wesley, Reading, 1960), Chap. 15.

[16] We suggest that the excitation of several electron-hole pairs leads to a significant screening of electron-hole correlations, and therefore the carrier system in the dots will correspond to an electron-hole plasma. This is supported by the good overall agreement with the single particle calculations.



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