Photoluminescence from a Single GaAs/AlGaAs Quantum Dot

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Isolated single quantum dots of different size have been fabricated by laser-induced local interdiffusion of a GaAs/AlGaAs quantum-well structure. Microscopic photoluminescence (PL) reveals a splitting and a blueshift, which depend systematically on dot size. The distinct PL peaks separated in energy by up to 10 meV are attributed to recombination between zero-dimensional (0D) electron and hole states. Complete quantization and inherent exclusion of inhomogeneous broadening in a single dot structure cause PL linewidths below 0.5 meV. The strength of the higher-energy transitions indicates the slowed energy relaxation theoretically predicted for 0D systems.

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One- and zero-dimensional semiconductor structures have become objects of extensive research activity recently [1–6]. Although techniques of lateral patterning are going to be greatly improved, inhomogeneity within arrays of low-dimensional systems still cause important line broadening. This especially applies to optical spectroscopy of the band-gap recombination which is strongly affected by fluctuations in composition and in structure size.

In this Letter we report on microscopic spectroscopy of a single quantum dot. The experimental results obtained this way should not be affected by inhomogeneous broadening. Local potential fluctuations in a single dot may modify the quantization energy of discrete atomic-like levels but should not cause inhomogeneous line broadening. The confining barriers have been created by focused-laser-beam- (FLB-) induced local interdiffusion of a GaAs/AlGaAs single-quantum-well structure. Al/Ga interdiffusion results in an increase of the effective quantum-well band gap. The level splitting and blueshift observed in photoluminescence (PL) from a series of dots of different size are qualitatively understood by considering the lateral modulation of quantum-well interdiffusion. The PL lines reveal energy splittings up to 10 meV, which are attributed to 0D electron-hole transitions. Linewidths below 0.5 meV are observed at low-excitation-power conditions. The relative PL intensity of excited levels increases with the level splitting. A simple model based on calculations of acoustical phonon scattering of electrons and holes shows that this result can be understood in terms of a strong decrease of the electron relaxation rate with increasing lateral confinement. This effect is expected to limit the PL efficiency of 0D systems in an intrinsic manner [7].

A schematic cross section of a quantum dot structure discussed here is given in the inset of Fig. 1. The starting material is an undoped single-quantum-well sample grown by molecular beam epitaxy (MBE). An $L=30$ Å thick GaAs quantum well is embedded in $Al_{0.35}Ga_{0.65}As$ layers of 200-Å width. On top of the 100-Å GaAs cap layer a Si_3N_4 film of 1000-Å thickness has been plasma deposited in order to prevent oxidation and ablation of the surface layer during laser processing. A band-gap modulation is produced by locally heating the sample by an Ar^+ laser beam focused to a spherical spot with a Gaussian intensity profile of 500-nm width at half maximum ($\lambda=514.5$ nm; $P=5.5$ mW). An external laser power stabilizer and an autofocus system are employed to keep the laser intensity at the sample surface constant. At temperatures of about 1000 °C rapid thermal interdiffusion of the topmost sample layers takes place under-

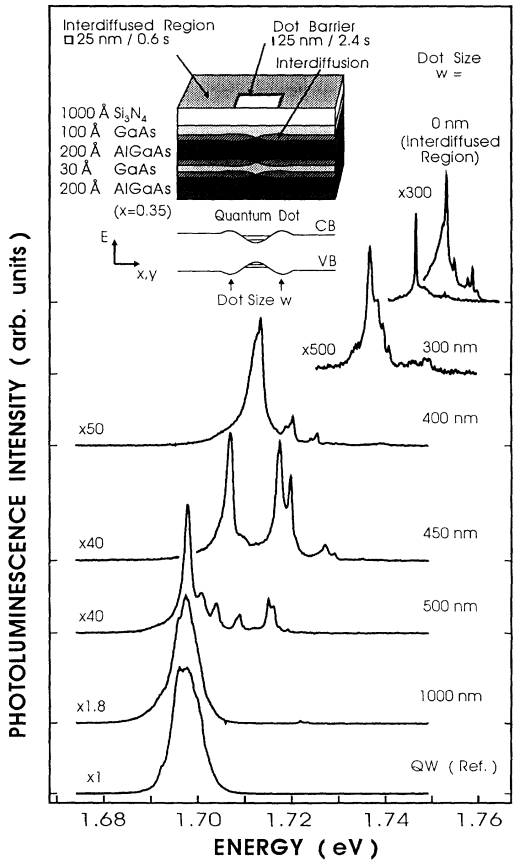

FIG. 1. PL spectra of single dot structures of various size w. Laser excitation was at $E_{exc}=1.96$ eV with $P=1$ μW focused to a spot of size 1.5 μm. Inset: A schematic diagram of a dot structure fabricated by FLB-induced interdiffusion of a 30-Å-thick $GaAs/Al_{0.35}Ga_{0.65}As$ single quantum well.

neath the laser spot center. This has been monitored by *in situ* Raman spectroscopy of the AlGaAs and GaAs phonon modes [8]. Thermal interdiffusion of Al and Ga with an activation energy of about 5 eV results in lateral potential barriers which are about 5 times steeper than the laser intensity profile [9,10]. A dot structure is defined by drawing a square frame of size w with the focused laser beam. This is done by moving the sample on an x-y-z translation stage of 10-nm accuracy with a time delay of 2.4 s after each 25-nm-sized step of motion. An area of 6 μm×6 μm surrounding the dot has been interdiffused homogeneously with a time delay of 0.6 s at each 25-nm grid position. The intended band-gap modulation of the buried dot structure is given schematically in the lower part of the inset. PL and PL excitation (PLE) measurements with a spatial resolution of about 1.5 μm were performed at a sample temperature $T=5$ K using a cold-finger He flow cryostat at the microscope stage. Excitation was done by a HeNe laser or a tunable Ti-sapphire laser at a typical power of 1 μW at the sample. The spectra were recorded by a triple-grating Raman spectrometer with a Si-diode multichannel detector or with a cooled photomultiplier tube.

Dot structures with geometrical widths ranging from 250 to 2000 nm have been written at nominally constant FLB processing conditions. Some typical PL spectra are shown in Fig. 1. At large dot size $w \approx 1000$ nm the PL line resembles the luminescence of the as-grown quantum well with a linewidth of about 7 meV (Ref. in Fig. 1). By reducing w to 450 nm the PL exhibits a more and more pronounced peak structure and shifts slightly to higher energies. Below 450 nm the blueshift continues to increase but the peak splitting decreases. The dependence of PL peak energies on dot size is summarized in Fig. 2. It can be understood considering the lateral carrier confinement and the change of the effective band gap in the dot center, which are both induced by the lateral modulation of quantum-well interdiffusion. In large structures a wide noninterdiffused area causes negligible quantization and PL energy shift. At $w=450$ nm there is maximum energy splitting of the equidistant PL main

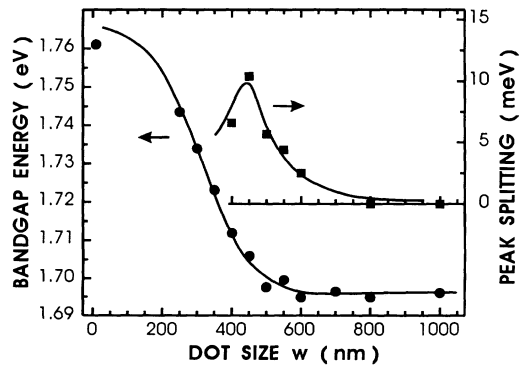

FIG. 2. Energy splitting of predominant PL peaks and minimum PL peak energy vs the geometrical dot size w. The solid lines are just a guide to the eye.

peaks $\Delta E=10$ meV, and a blueshift of comparable size is observed. This splitting and blueshift at $w=450$ nm compare well to the transition energies of localized electron and hole levels with equal lateral quantum number $n_e=n_h=1,2,3$, which have been calculated from the FLB-induced quantum-well interdiffusion as described below. A barely interdiffused dot center with steep lateral potential barriers is realized in this case. The blueshift of the ground-state energy is mainly attributed to a ground-state quantization energy of about 10 meV, while a shift of only a few meV is caused by interdiffusion at the dot center. At small dot size $w \approx 300$ nm, however, most of the energy shift of more than 40 meV is due to interdiffusion at the dot center. The lateral quantization energies are reduced to a few meV by the flattening of the confining potential. Within the interdiffused regions around the dots detectable PL (spectra $w=0$ nm in Fig. 1) is observed only at some locations, which are attributed to local band-gap minima and defects. The maximum blueshift of about 70 meV is an estimate of the height of band-gap modulation. This corresponds roughly to the average band-gap energy of the topmost totally intermixed layers. Nonradiative recombination centers generated during laser processing are expected to reduce the PL efficiency within that strongly interdiffused area. The much higher efficiency and smaller blueshift of PL from the active dot structures compared to that from the surrounding region allows spectroscopy of isolated dots at low excitation such that the estimated occupation of the dot is smaller than one e-h pair on a time average.

PL splitting and blueshift are well described by a simple model of local quantum-well interdiffusion based on our laser processing and sample parameters. An error-function-like profile for the Al content is assumed at the interdiffused interfaces in the growth direction z [1]. Solving the steady-state heat conduction equation for an $Al_{0.35}Ga_{0.65}As$ crystal with a heat source given by the absorbed FLB, we have calculated the modulation of Al content within the quantum well [10,11]. Separating the wave function along z we obtain similar lateral modulations of the effective conduction and valence band edges, which are nearly isotropic and parabolic near the dot center. Neglecting Coulomb interaction and interband coupling this confinement results in equidistant dot levels $n_{e,h}=1,2,3$ of degeneracy $n_{e,h}$, neglecting spin. With a realistic value for the laser spot size we get lateral quantization energies of 6 and 4 meV for electrons ($m_e=0.07m_0$) and heavy holes ($m_{hh}=0.11m_0$), respectively, at a dot size $w=450$ nm.

The observation of transitions between excited states is a remarkable result. PL spectra of three-, two-, and one-dimensional III-V semiconductor structures usually show only one strong transition corresponding to the ground state. It has been shown theoretically that in 0D semiconductor systems the relaxation by longitudinal acoustic (LA) phonon scattering decreases strongly when the separation of the discrete electronic levels increases above a

TABLE I. Calculated LA phonon scattering rates of electrons and holes, τ_e^{-1} and τ_h^{-1}, respectively, and relative intensity (I_2/I_1) of the first excited to the fundamental PL peak for the quantum dots of geometrical size w.

w (nm)	τ_e^{-1} (ns^{-1})	τ_h^{-1} (ns^{-1})	I_2/I_1
400	1.8	28	0.08
450	0.015	7.8	2.0
500	3.5	31	0.07

threshold E_t [12]. Although conservation of electron-phonon momentum during scattering is relaxed due to the confinement, above E_t the energy conservation requires the emission of LA phonons with a wave vector too large to be accommodated by the electrons. The threshold energy E_t, given by $E_t = \hbar c_s 2\pi/L$, only depends on the sound velocity c_s (3700 m/s for GaAs [13]) and the smallest dimension of the quantum dot $L = 30$ Å. Throughout our series of quantum dots $E_t = 5.1$ meV is constant. These single-particle results should not be changed qualitatively by electron-hole interaction [14]. Let us compare the spectra for $w = 400$, 450, and 500 nm of Fig. 1. From the experimental peak separation we deduce electron and hole level separations ΔE_e and ΔE_h, assuming $\Delta E_h/\Delta E_e = (m_e/m_{hh})^{1/2}$. We calculate the rate of LA phonon scattering from the first excited to the fundamental level for electrons (τ_e^{-1}) and holes (τ_h^{-1}). Details of the theory are given in Ref. [12]. Table I shows that in the 450-nm sample the scattering rate of electrons is 2 orders of magnitude smaller than the rates obtained for $w = 400$ and $w = 500$ nm. This result is expected from the above discussion since in the 450-nm sample $\Delta E_e > E_t$ while ΔE_e and ΔE_h are below E_t in the other cases. This already suggests that the experimental observation of a much stronger higher-energy transition in the 450-nm sample is related to a decrease of electron relaxation with increasing level spacing. More quantitatively we have studied the influence of the relaxation rates on the optical spectra by solving the rate equations of a four-level model (ground state and first excited degenerate state of electrons and holes $e1, e2$ and $h1, h2$, respectively). It includes radiative recombination $e1 \rightarrow h1$ and $e2 \rightarrow h2$ with an assumed lifetime $\tau_r = 250$ ps, nonradiative loss from all levels with a lifetime $\tau_{nr} = 10$ ns [7], and the calculated relaxation rates τ_e^{-1} and τ_h^{-1}. Table I shows that the resulting values for the relative intensities of the first excited to the fundamental transition (I_2/I_1) explain the experimentally observed intensity ratios. The presence of strong high-energy transitions at large peak spacing is thus expected to be due to a slowed electron relaxation. Together with the systematic dependence of the transition energies on the dot size w these experimental results strongly indicate that the observed PL splitting reflects the discrete energy spectra of a 0D system and is not caused by potential fluctuations or defects.

In the following we will focus on the dot $w = 450$ nm, which exhibits the maximum peak splitting $\Delta E = 10$ meV. PLE spectra are shown in Fig. 3. They strongly contrast to the PLE spectrum of the as-grown quantum well, which reveals a PLE heavy-hole exciton peak Stokes shifted by 8 meV and a light-hole peak at 34 meV higher energy. Detecting the intensity of the 0D ground-state recombination (PLE 1), absorption peaks with an equidistant energy separation of about 2 meV are observed close to it. The broad resonance at energy $E = 1.718$ eV corresponds to the PL peak of the first excited electron level. At higher energy a continuous increase of intensity to a constant value above $E = 1.75$ eV with weak shoulders implies an increasing number of absorption channels. Absorption peaks in this energy range are well resolved, detecting the intensity of the $n_e = 2$ double peak (PLE 2,3). This again indicates that electrons generated in excited levels are hindered from fast relaxation to the

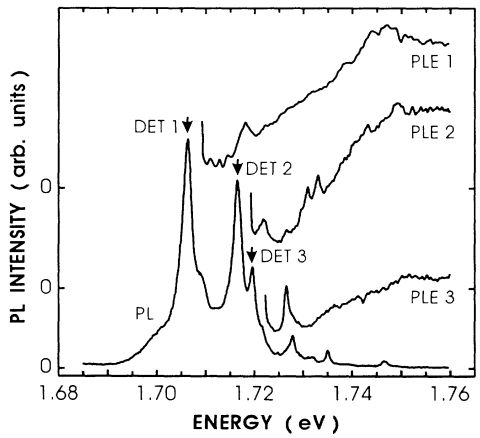

FIG. 3. PL and PL excitation spectra of the dot $w = 450$ nm. The spectra PLE 1,2 are shifted vertically for clarity. The detection energies are indicted by arrows. The spectral resolution was 0.5 meV.

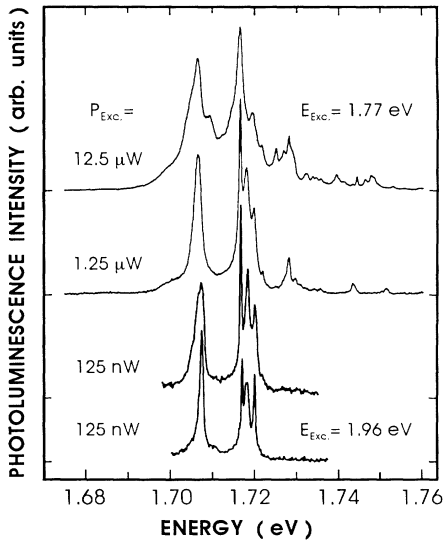

FIG. 4. PL spectra from the dot of size $w = 450$ nm on an intensity scale normalized by the different laser excitation power. The photon energy was $E_{exc} = 1.77$ eV, except for the bottom spectrum with $E_{exc} = 1.96$ eV.

ground state. PLE of the dot recovers the peaked density of states observed in PL modified by details of the carrier relaxation and recombination [7]. Radiative recombination in excited levels is pointed out by the high-energy PL peaks.

In Fig. 4 PL spectra are shown for various intensities normalized by their different excitation power. The photon energy $E_{exc}=1.77$ eV used in the upper spectra corresponds to an excitation near the top of the confining potential. At lowest excitation $P=125$ mW the PL reveals a single peak of the ground-state recombination at $E=1.707$ eV and three distinct peaks at about $E=1.72$ eV. The intensity of these two main features is approximately proportional to excitation power. No substantial saturation effects on the optical transitions are observed. With increasing power density the transitions at higher energy increase in efficiency concomitant with the appearance of additional shoulders and small peaks in between the predominant PL peaks. The PL lines broaden from widths of about 0.5 meV up to about 3 meV. The spectral widths are further reduced, exciting the dot in the continuum at energy $E_{exc}=1.96$ eV (lowest spectrum of Fig. 3). The width reduces to below 1 meV for the fundamental transition and below 0.5 meV for two of the peaks at 1.72 eV.

Within our simple model of noninteracting electrons and holes we attribute the predominant PL peak structure to quantum-number-conserving transitions between successive dot levels. The PLE peaks as well as the fine structure and shoulders in between the PL main peaks, which are both spaced by about 2 meV, may imply quantum-number-nonconserving transitions into consecutive hole levels. The deviation of this hole confinement energy from the calculated value of 4 meV might be due to a modified interdiffusion profile in growth direction and a subsequently different confinement of electrons and holes. Optical selection rules theoretically derived for symmetrical potentials can be weakened in our dot structures due to width fluctuations of the as-grown quantum well or a nonuniform lateral potential built up by interdiffusion. Along this line, the fine structure observed at photon energies about 1.72 eV is tentatively attributed to a splitting of the twofold-degenerate $n_e=2$ electron level and to the $n_h=1,2$ hole levels.

The spectral width of individual PL peaks is considered to be without an inhomogeneous contribution in a single 0D quantum structure. In our experiment negligible lattice heating by the laser spot excludes strong broadening by phonon scattering. The increase of linewidth with excitation power in Fig. 4 suggests a mechanism which depends on the occupation of the dot levels. This is supported by the increased spectral width observed at resonant excitation of the upper dot levels compared to excitation in the continuum (two bottommost spectra in Fig. 4), assuming a higher average occupation of excited levels in the former case. Carrier-carrier scattering and Coulombic shifts of transition energy induced by a temporally fluctuating occupation of dot levels are possible candidates to explain the line broadening in our time-averaged spectra.

In conclusion, we have fabricated quantum dots of high optical quality by laser-induced thermal interdiffusion of a quantum-well structure. The first PL and PLE spectra from a single quantum dot are presented. As a result of the absence of inhomogeneous broadening this opens the unique possibility of studying intrinsic optical properties of artificial 0D systems. The spectra show sharp transitions between 0D electron and hole states. The intense PL from excited 0D levels experimentally observed is understood in terms of a strongly reduced electron relaxation.

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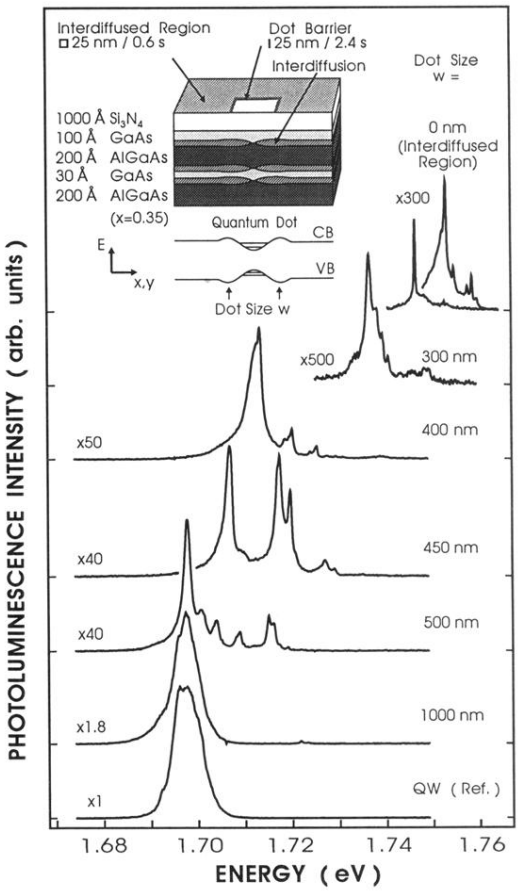

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